UT-BATTELLE

Fundamentals of Radiation Damage: Fun with Vacancies and Interstitials

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ATR NSUF Summer School

Fuels and Materials Course

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The primary objective of this talk is to explore the basics of radiation damage and basic effects.

Outline

- Introduction to Radiation Damage
- The “5-scourges of radiation damage!”
  - Diffusion-related radiation degradation
  - Clustering-related radiation degradation
- Other forms of degradation

What should you expect to take away from this lecture?

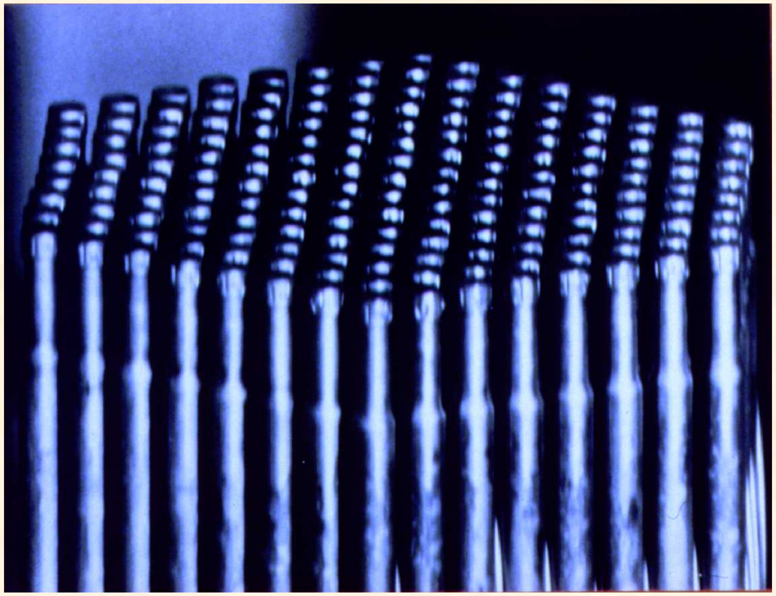
- Basic understanding of neutron interactions with crystalline materials
- An appreciation for the exciting lives of vacancies and interstitials
- A survey of modes of degradation common to LWR and fast reactors
- No equations

Fundamentals of Radiation Damage

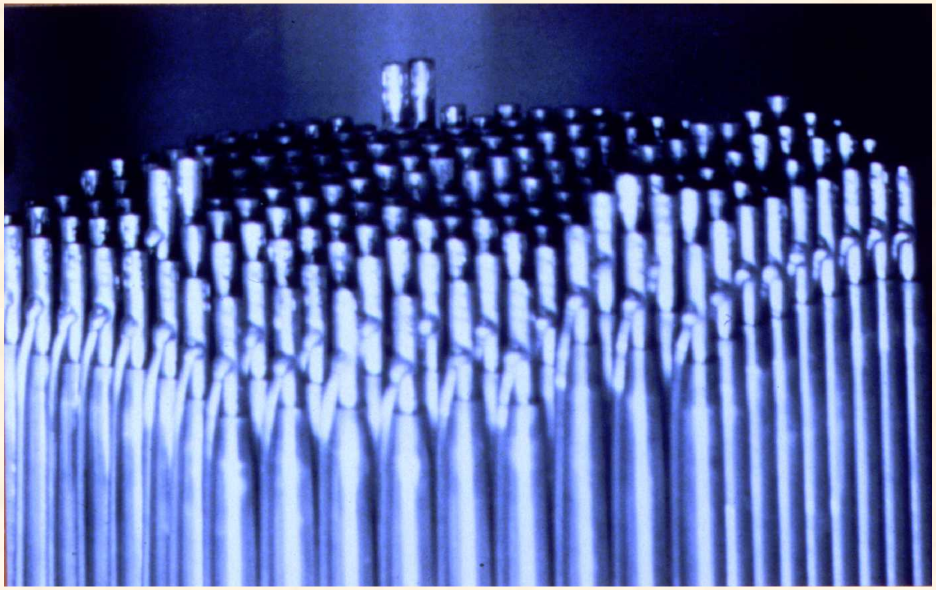
Motivation: Why do we care about radiation damage?

Swelling can create tolerance problems

FFTF Fuel Pin Bundles



HT-9, no swelling



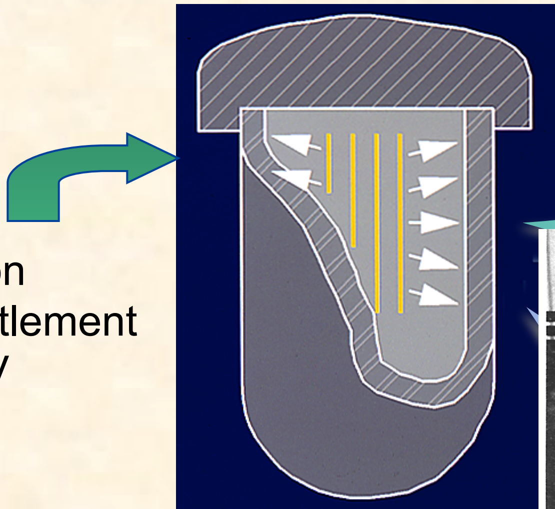
316-Ti stainless, swelling

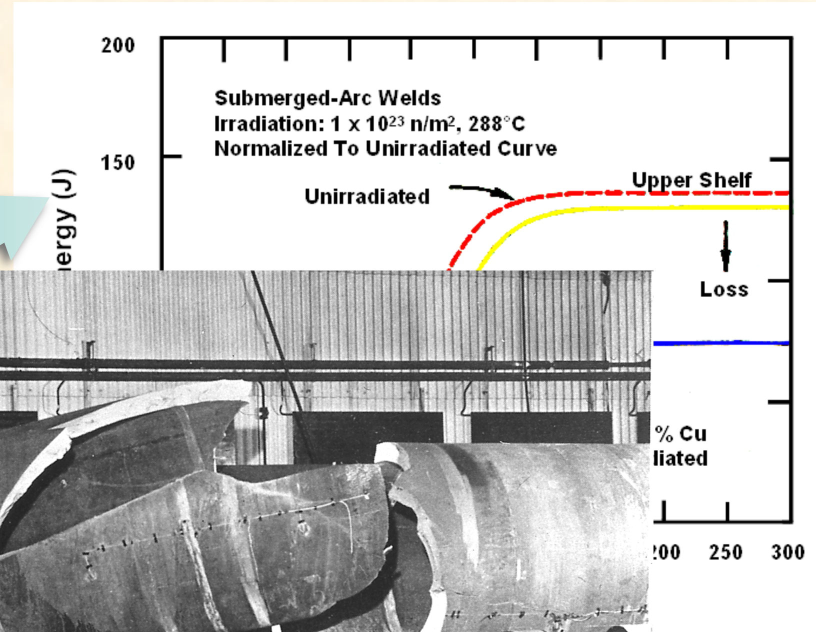
Source: F. Garner

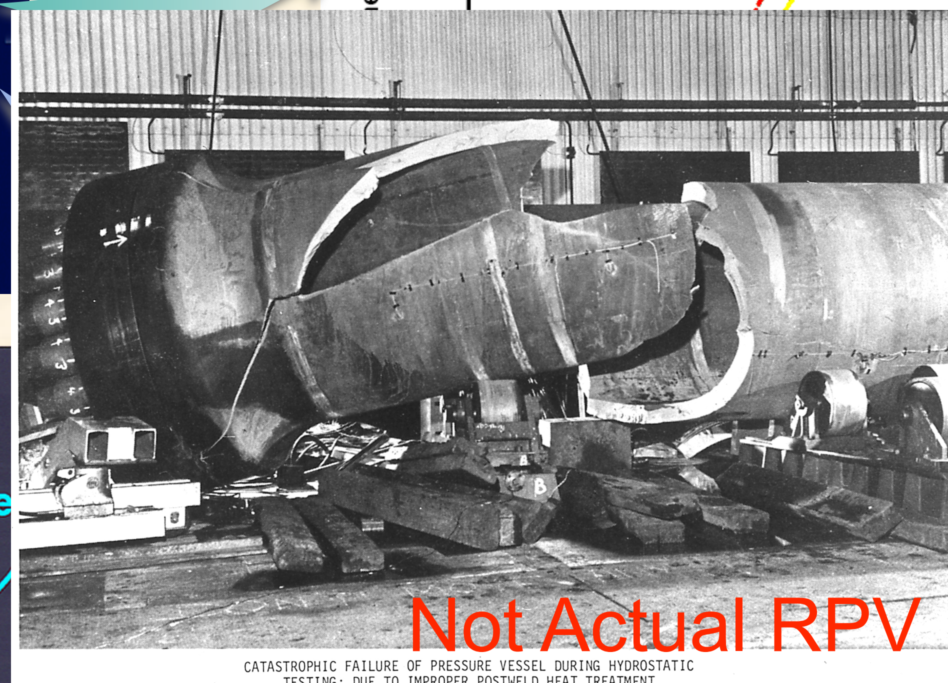
Reactor Vessel Integrity Experiences Potential Degrading Effects of Neutron Irradiation

Irradiation Causes Ductile/Brittle Transition Temperature Shift and Upper Shelf Energy Loss — Copper Increases The Effect

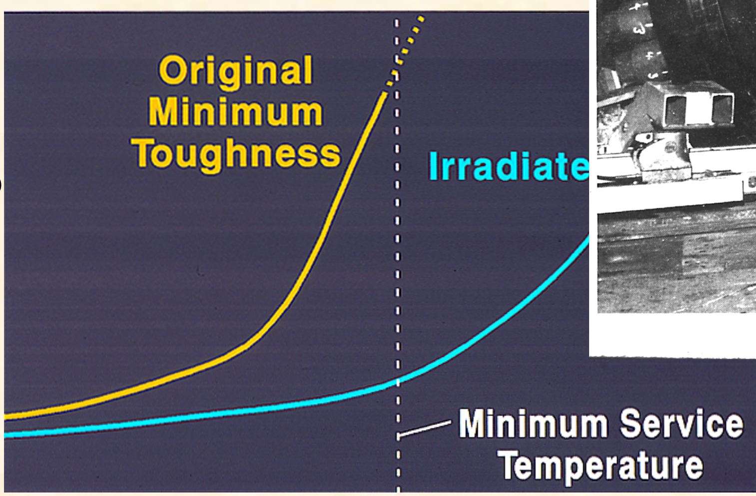
Neutron Embrittlement of RPV







Fracture Toughness

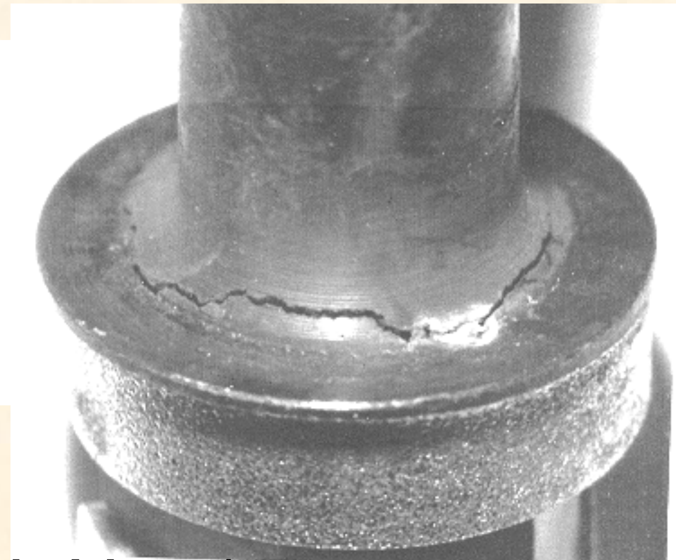


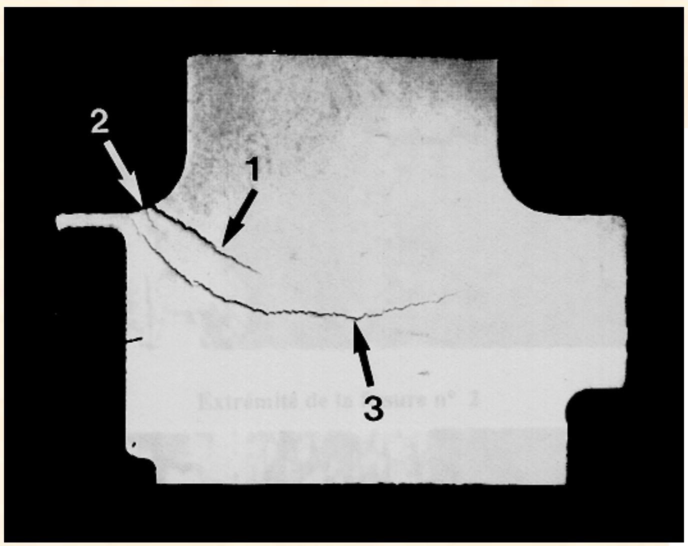
Temperature

…d …uctures: …ates and Matrix Damage

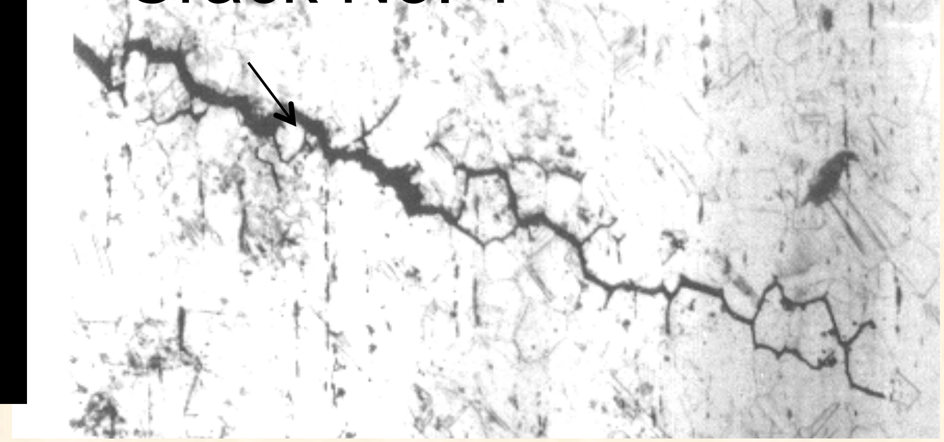
Source: R. Nanstad

IASCC affects baffle former bolts in PWRs





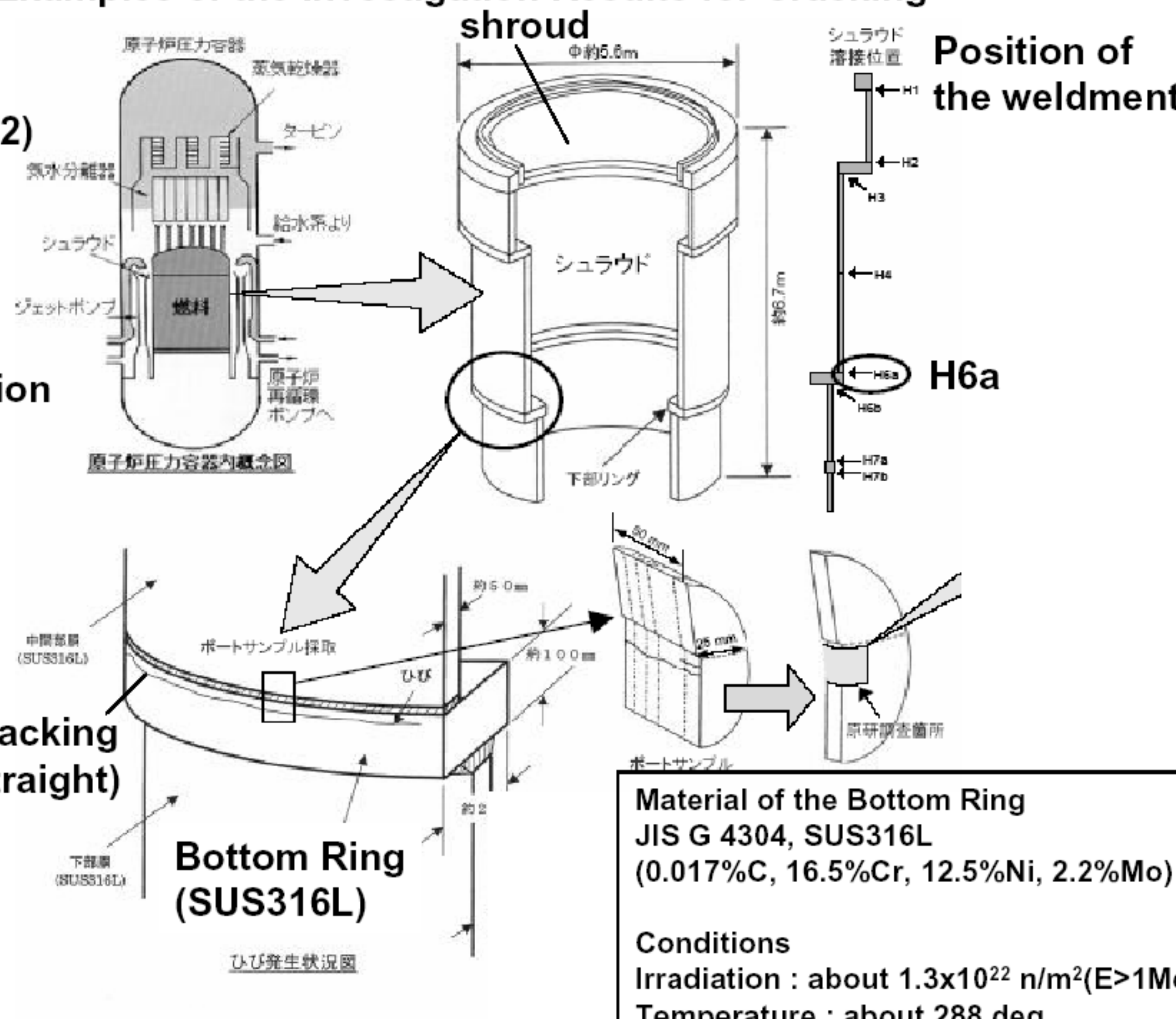
Crack No. 1



Source: G.S. Was

3. Research ~Examples of the Investigation Results for Cracking~

3.1 Shroud
(Unit3
-Fukushima2)



Material of the Bottom Ring
JIS G 4304, SUS316L
(0.017%C, 16.5%Cr, 12.5%Ni, 2.2%Mo)

Conditions
Irradiation : about 1.3×10^{22} n/m^2(E>1MeV)
Temperature : about 288 deg.
Dissolved Oxygen : about 250 ppb

Source: G.S. Was

Cracking of the Shroud (2F-3) ~ Microscopic observation ~

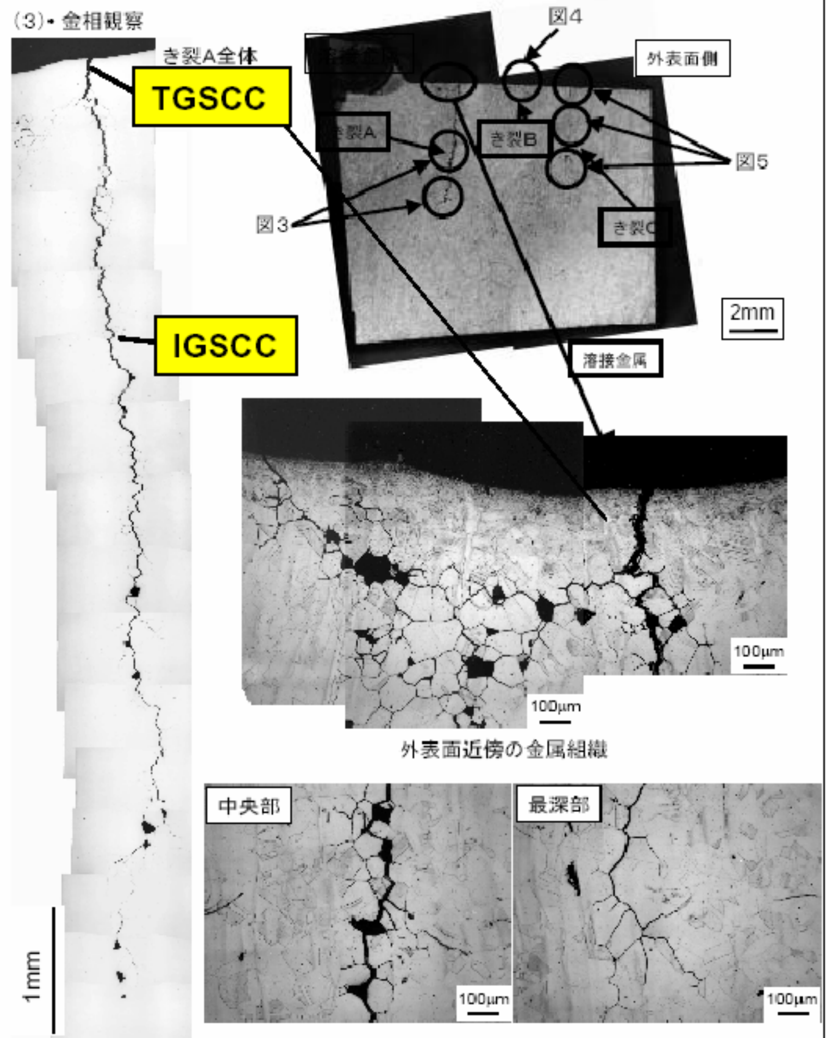


図3・き裂Aの全体（左図）及び中央部、最深部の金属組織（エッチング後）

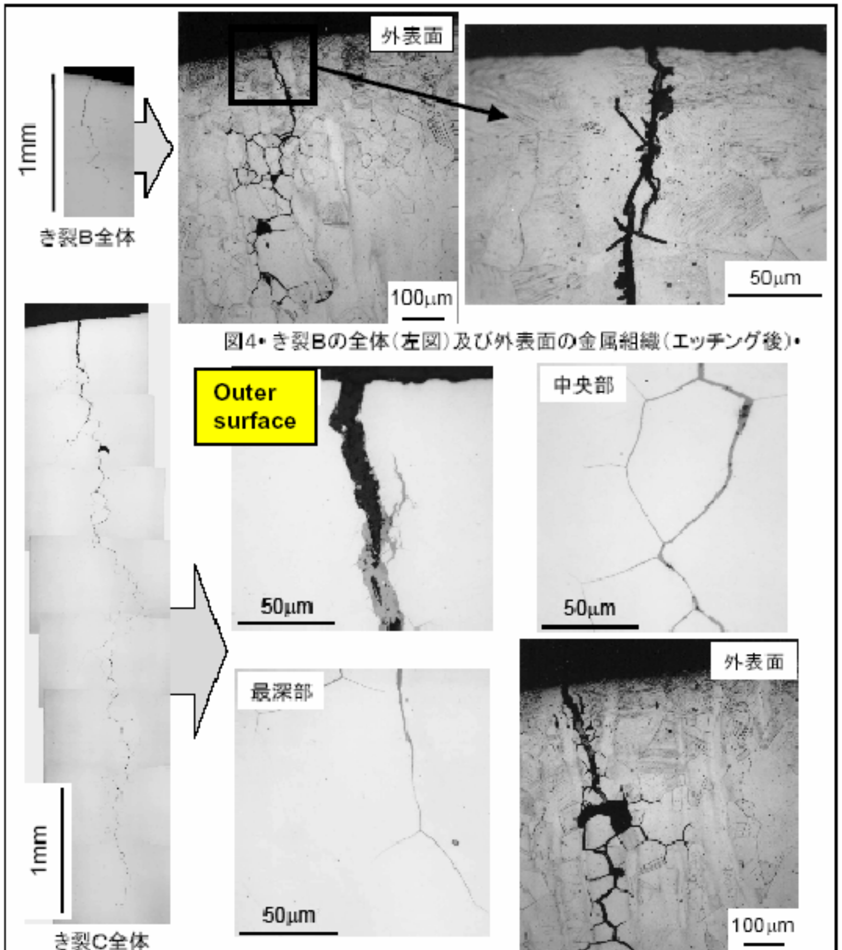


図5・き裂Cの全体（左図）、外表面、中央部及び先端部の金属組織（エッチング前後）

まとめ：・き裂は3箇所に観察され、いずれのき裂も加工組織部に粒内割れが見られ、
・・・・・・ それ以外では2次き裂を伴う粒界割れの様相を呈していた。
・・・・・・・き裂開口部の外表面近傍には加工組織が約200～300μm見られる。

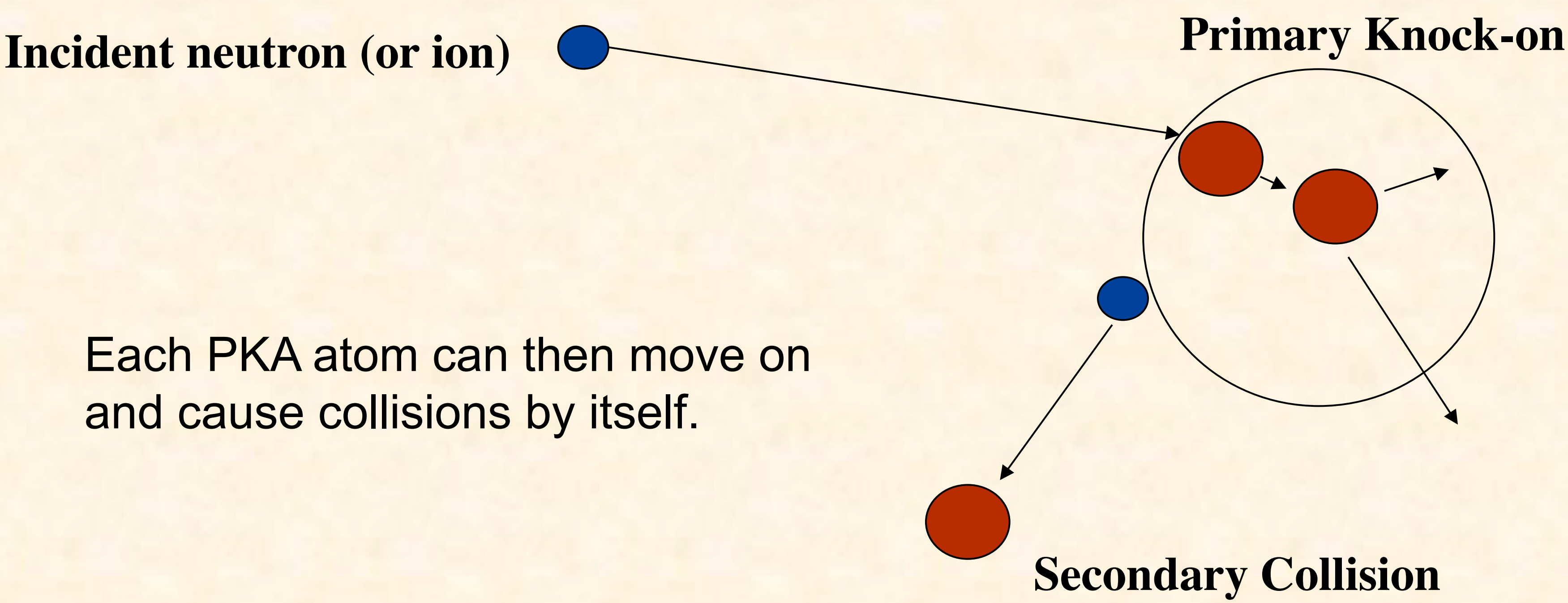
Source: G.S. Was

Fundamentals of Radiation Damage

The basics

Radiation Damage: the basics

- **All of radiation damage boils down to a common step: collisions between incoming neutrons and atoms in the crystal lattice!**



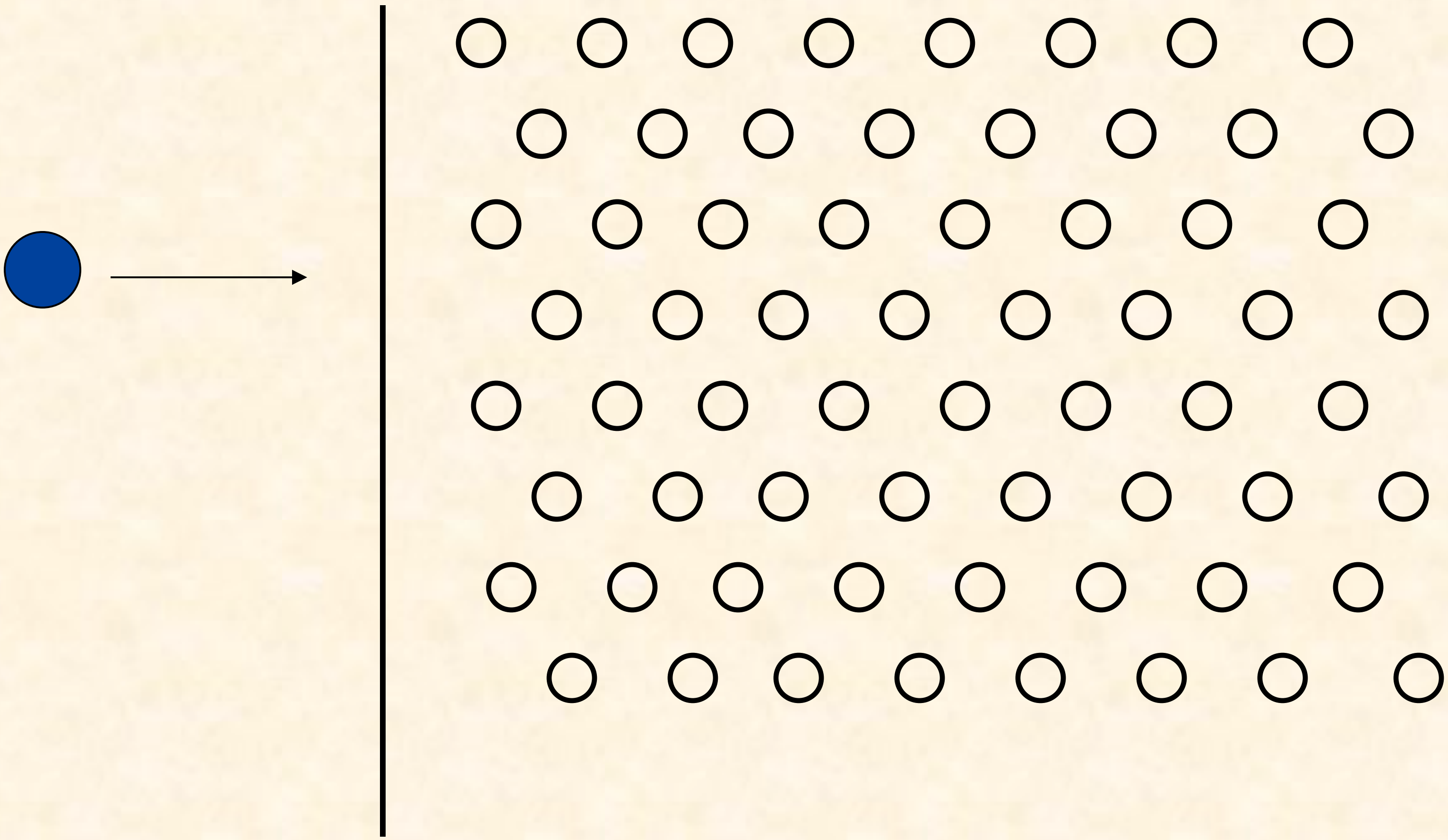
Primary Knockon Atoms are an important part of the damage process

- Each neutron/atom collision transfers energy. For neutrons, this number varies
  - Average kinetic energy for PKA in a fission reactor: 10 keV
  - Average kinetic energy for PKA in a fusion reactor: 50 keV

- As long as the energy of the PKA is above the energy to displace an atom (E_d ~ 40-50 eV), each PKA will transfer energy to other atoms in the crystal.

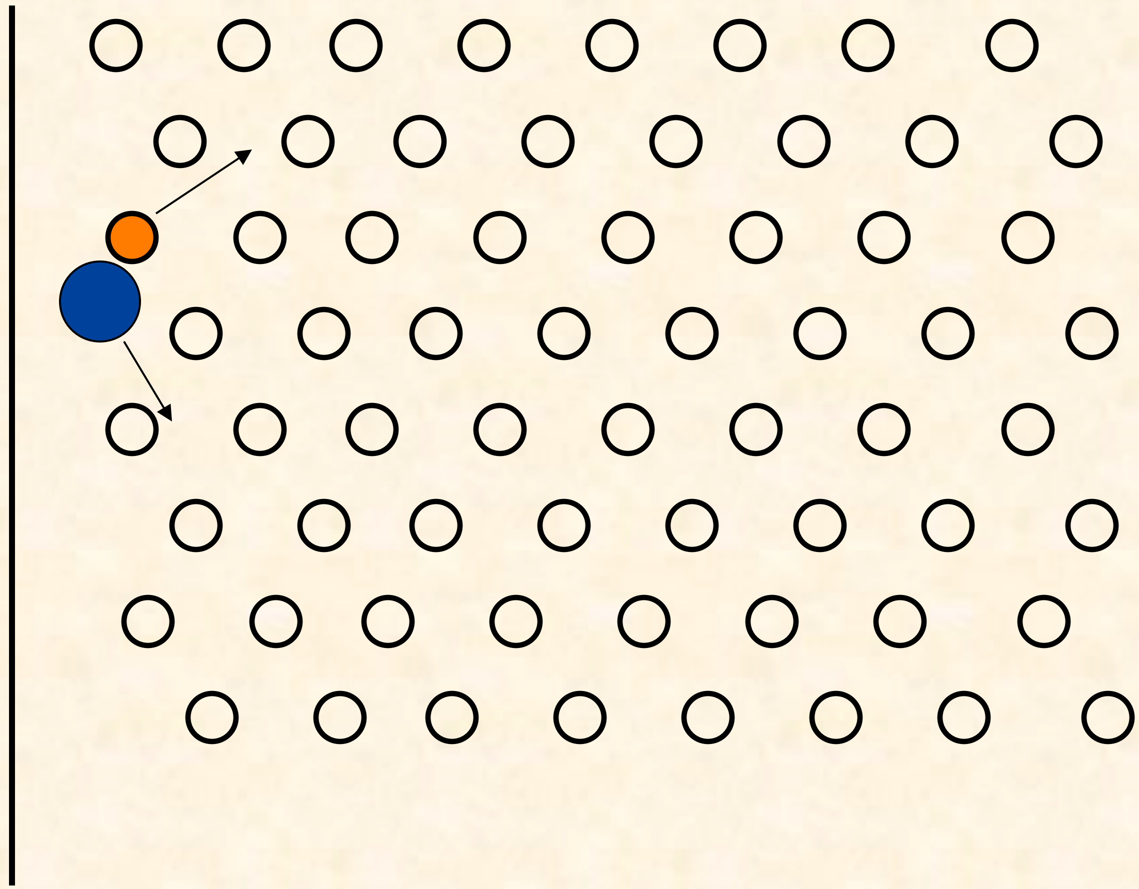
- This process continues creating a cascade.

Simple Picture



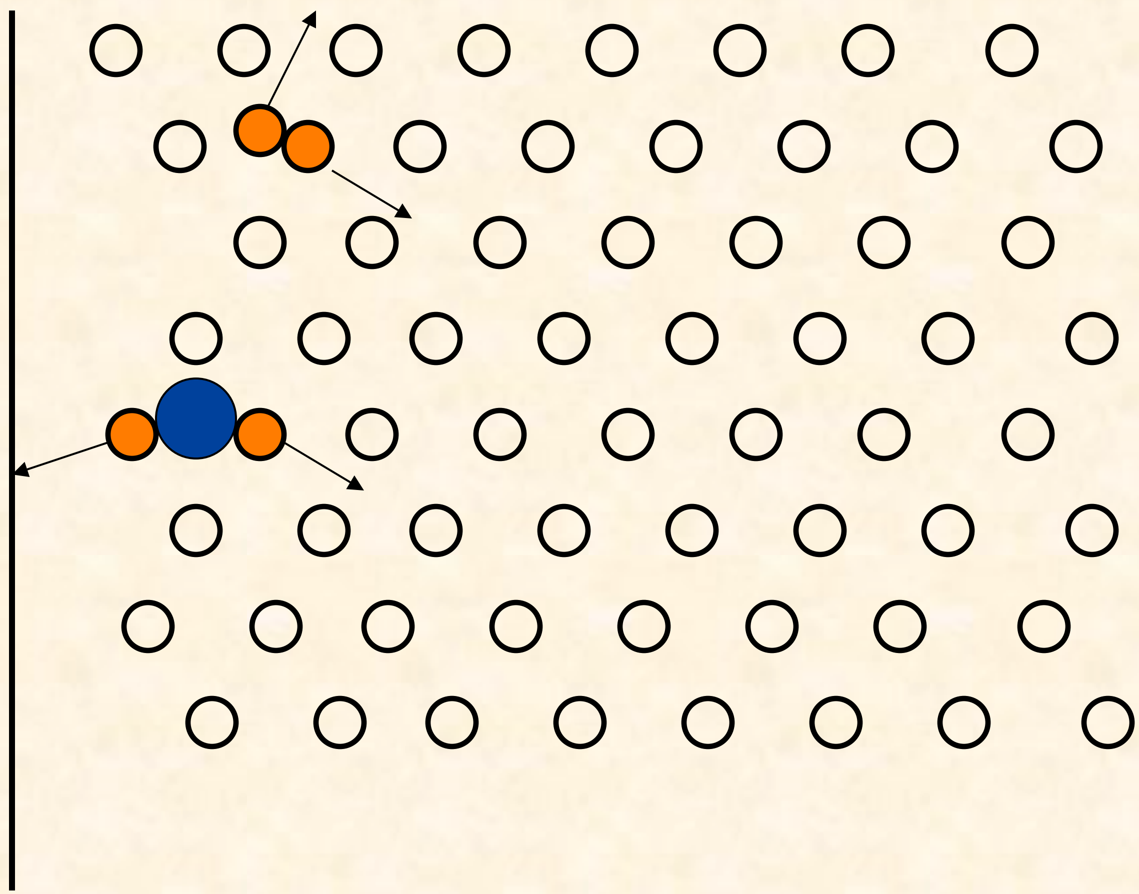
Source: T.R. Allen

Simple Picture



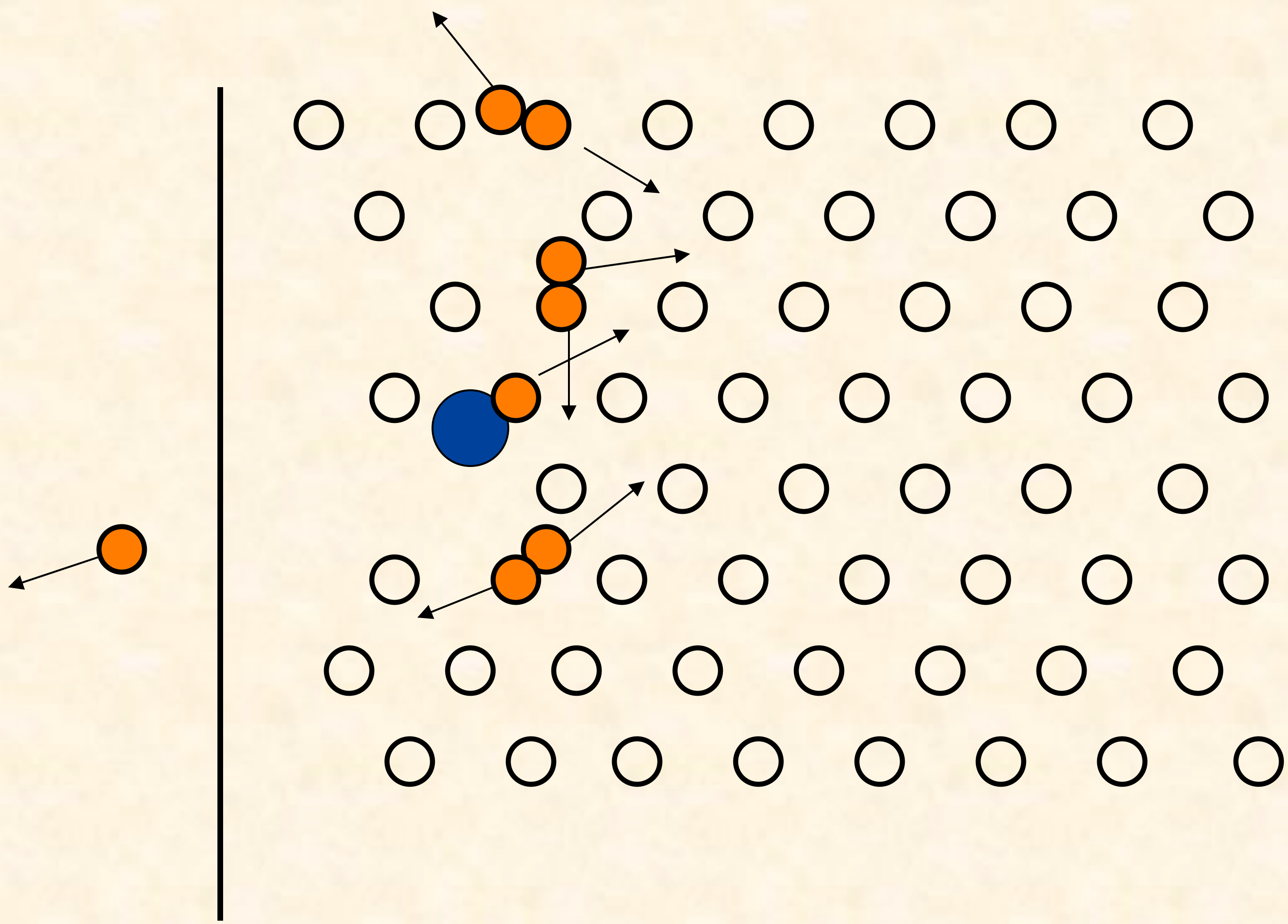
Source: T.R. Allen

Simple Picture



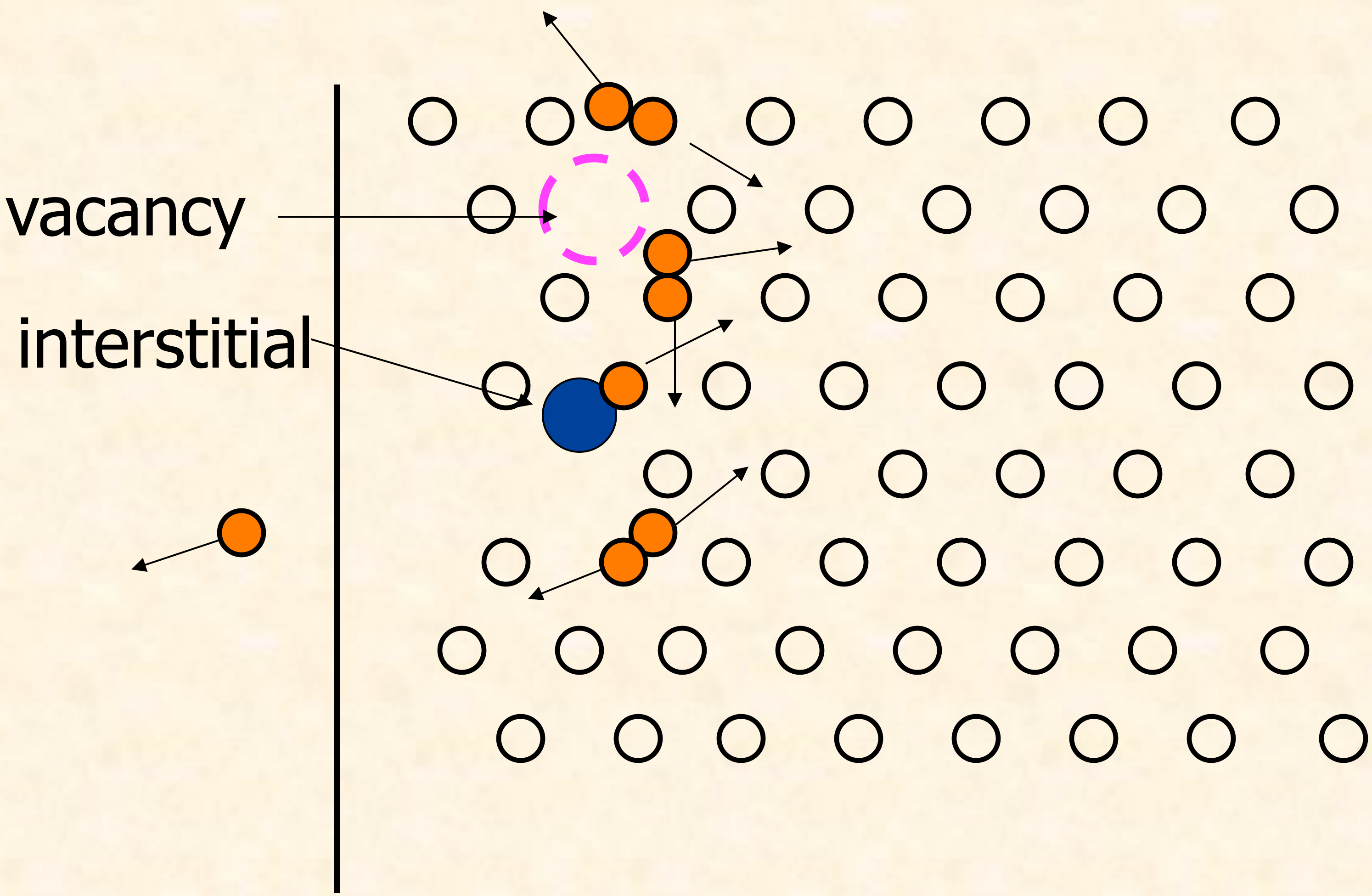
Source: T.R. Allen

Simple Picture



Source: T.R. Allen

Simple Picture



Source: T.R. Allen

Molecular dynamics provides an easier way to visualize cascades

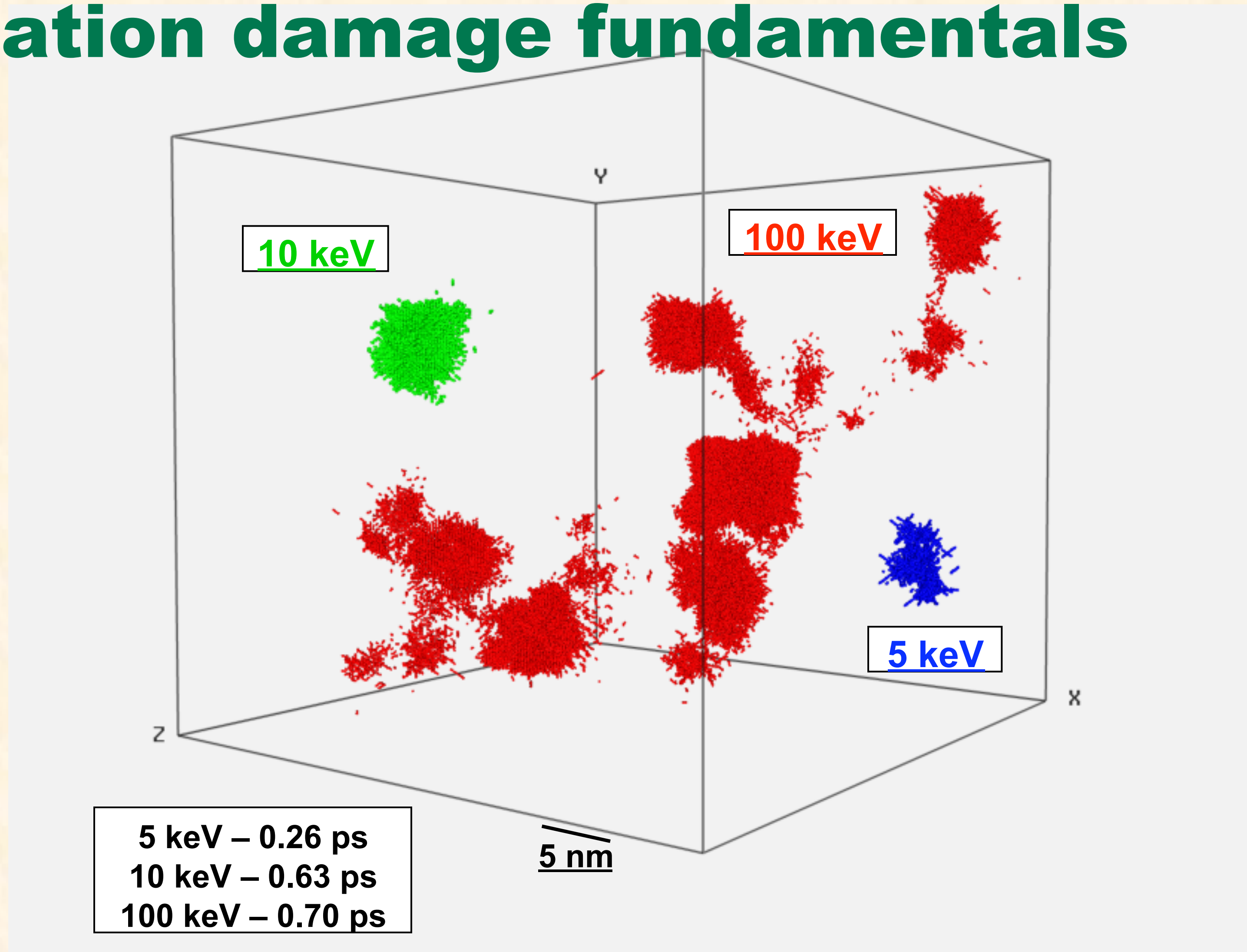
- While we can’t directly observe these interactions, we can model them.
- Molecular dynamic simulations have been invaluable in learning about these interactions.

Molecular dynamic simulations

- **MD movie:**

Source: R. Stoller

Molecular dynamics have provided considerable information on radiation damage fundamentals

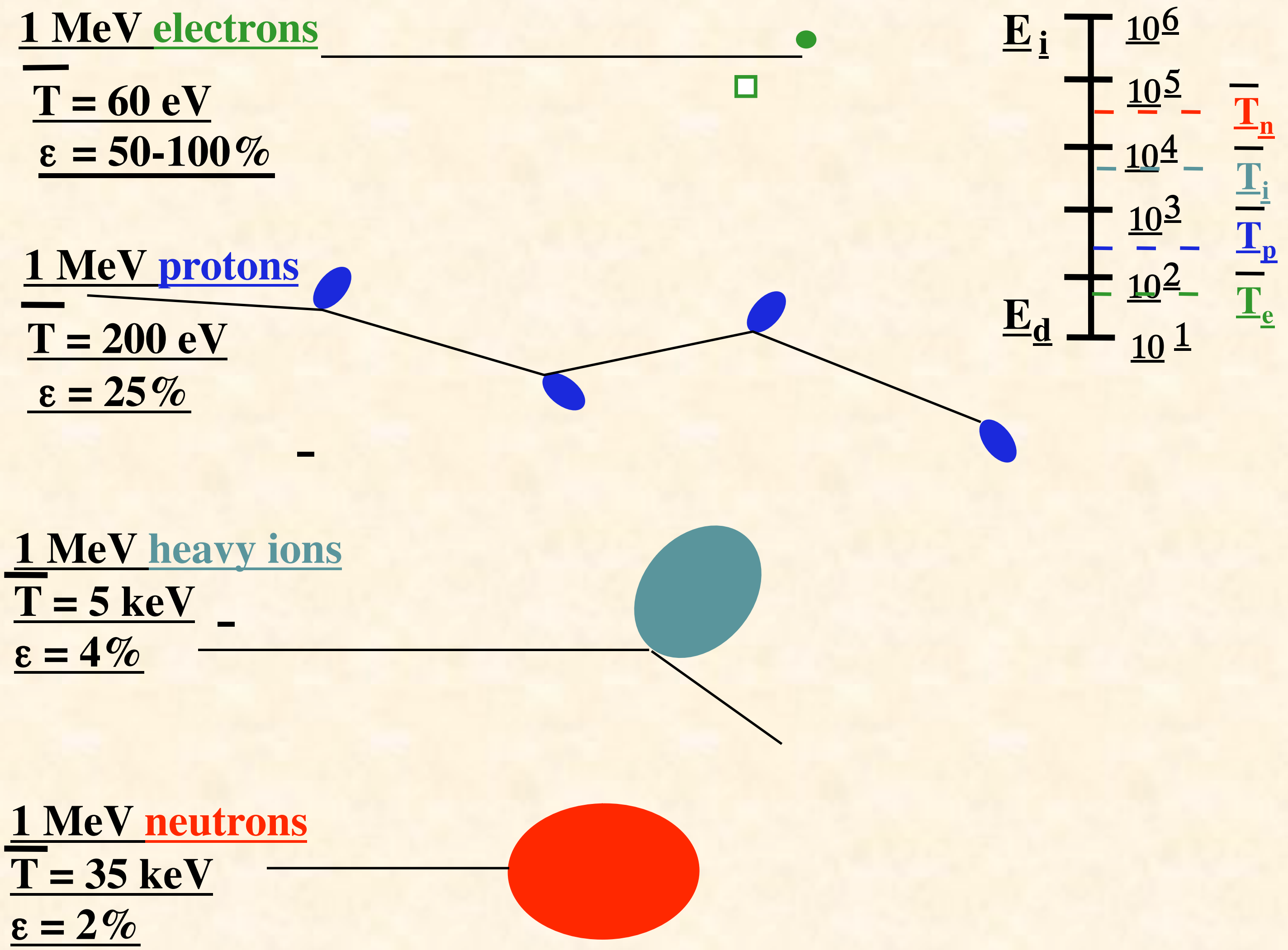


Source: R. Stoller

Dynamics of a displacement cascade

- The creation and subsequent decay of a displacement cascade consists of three overlapping stages.
  - Collisional phase ~ 10^{-13} s: All of the collisions associated with the PKA occur.
  - Cooling phase ~ 10^{-11} s: Annihilation, in-cascade clustering, replacement events occur.
  - Diffusional interactions phase > 10^{-11} s: Mobile defects interact in the lattice to form clusters or annihilate via recombination or at sinks.
- The total number of atoms displaced by each neutron depends on a number of factors:
  - Incoming particle (energy, direction)
  - Material (E_d, crystal structure)
  - Temperature

Ions also create cascades and defects



Source: G.S. Was

Damage is often normalized into “displacements per atom”

- **Displacement per atom (dpa) = average number of displacements of each lattice atom**

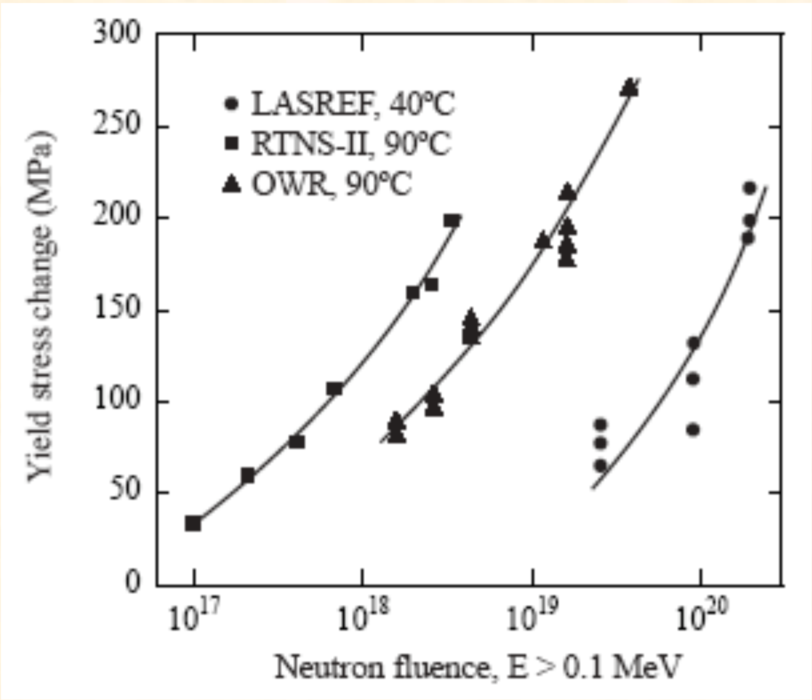
- **This calculated value is convenient as it can help normalize between different neutron spectra or even particles**

- **Rules of thumb:**
  - **0.7×10^{21} n/cm^2 (E>1 MeV) ≈ 1 dpa in SS**
  - **0.4×10^{21} n/cm^2 (E>1 MeV) ≈ 1 dpa in Zr**

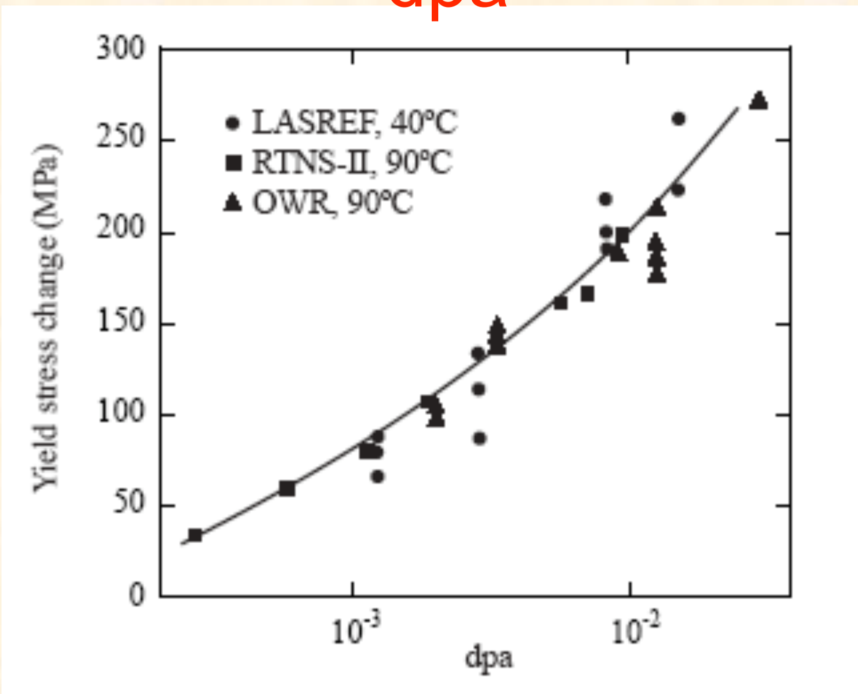
- **For a fast reactor core (1015 n/cm2/s), displacement rates of 10-6 dpa/s may be experienced. Each atom is displaced every ~12 days.**

Dpa can actually be validated as an accurate measure of damage

Fluence



dpa



Normalizing to DPA works!

Radiation-induced strengthening of 316 stainless steel in radically different neutron spectra (Heinisch and Martinez, 1986)

Fundamentals of Radiation Damage

How can vacancies and interstitials possible cause problems?

Radiation induced defects are produced in very large numbers

- Depending on material, temperature, and dose rate, excess vacancies and interstitials can be created at 4-6 orders of magnitude above thermal equilibrium.

- Defect populations in excess of equilibrium must be removed!

What happens when a system is far from equilibrium?

- That system tries to move back to equilibrium. But how?
- Defects can be removed in a number of ways
  - Recombination
  - Diffusion to surfaces
  - Diffusion to particles or precipitates
  - Diffusion to grain boundaries
  - Cluster together

Temperature and dose rate influence populations and their actions

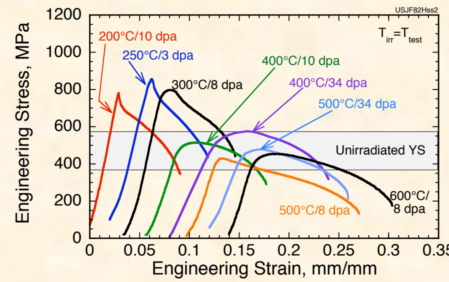
	Effect	Thermal Defect Population	Irrad. Defect Population	Diffusion	Typical Leading Process
Constant Dose Rate	Low Temperature	↓	—	↓	Clustering/ Recomb.
	Moderate Temperature	↑	—	↑	Diffusion
	High Temperature	↑	—	↑	Recomb./ Diffusion
Constant Temperature	Decrease Dose Rate	—	↓	↓	Diffusion/ Clustering
	Increase Dose Rate	—	↑	↑	Clustering/ Recomb.

Freely migrating defects and their actions are the key to radiation damage

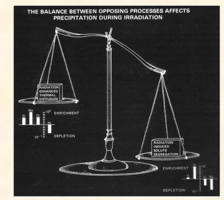
- Regardless of the nature of radiation (reactor, spectrum, particle), the surviving defects are the key for long-term material changes and properties
- Defects may
  - Recombine
  - Diffuse
  - Cluster
- The latter two possibilities will result in distinct changes.

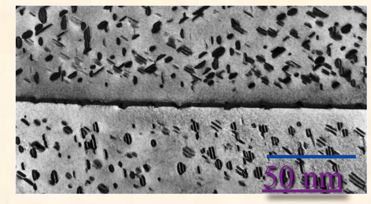
Radiation Damage can Produce Large Changes in Structural Materials

- **Radiation hardening and embrittlement ($<0.4\ T_M$, >0.1 dpa)**



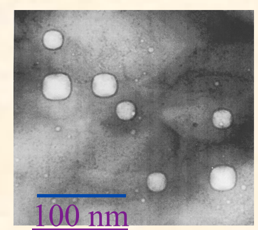
- **Phase instabilities from radiation-induced precipitation (0.3-$0.6\ T_M$, >10 dpa)**

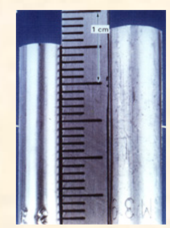




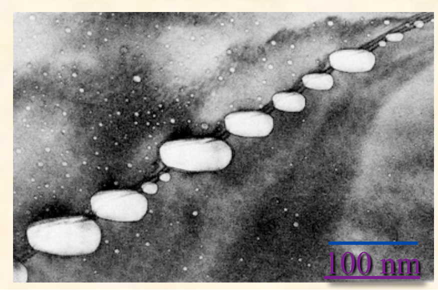
- **Irradiation creep ($<0.45\ T_M$, >10 dpa)**

- **Volumetric swelling from void formation (0.3-$0.6\ T_M$, >10 dpa)**





- **High temperature He embrittlement ($>0.5\ T_M$, >10 dpa)**

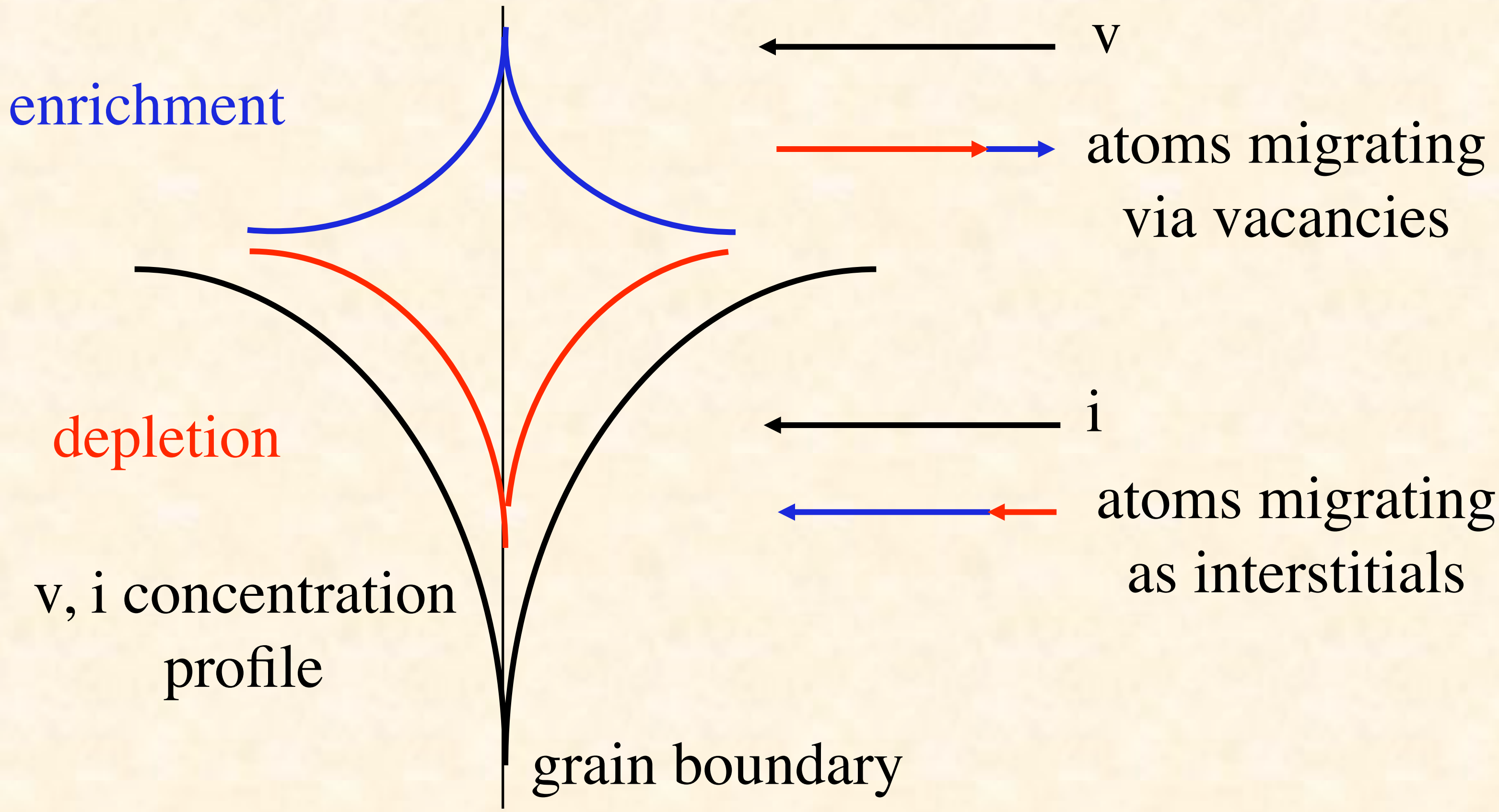


Source: S. Zinkle

Fundamentals of Radiation Damage

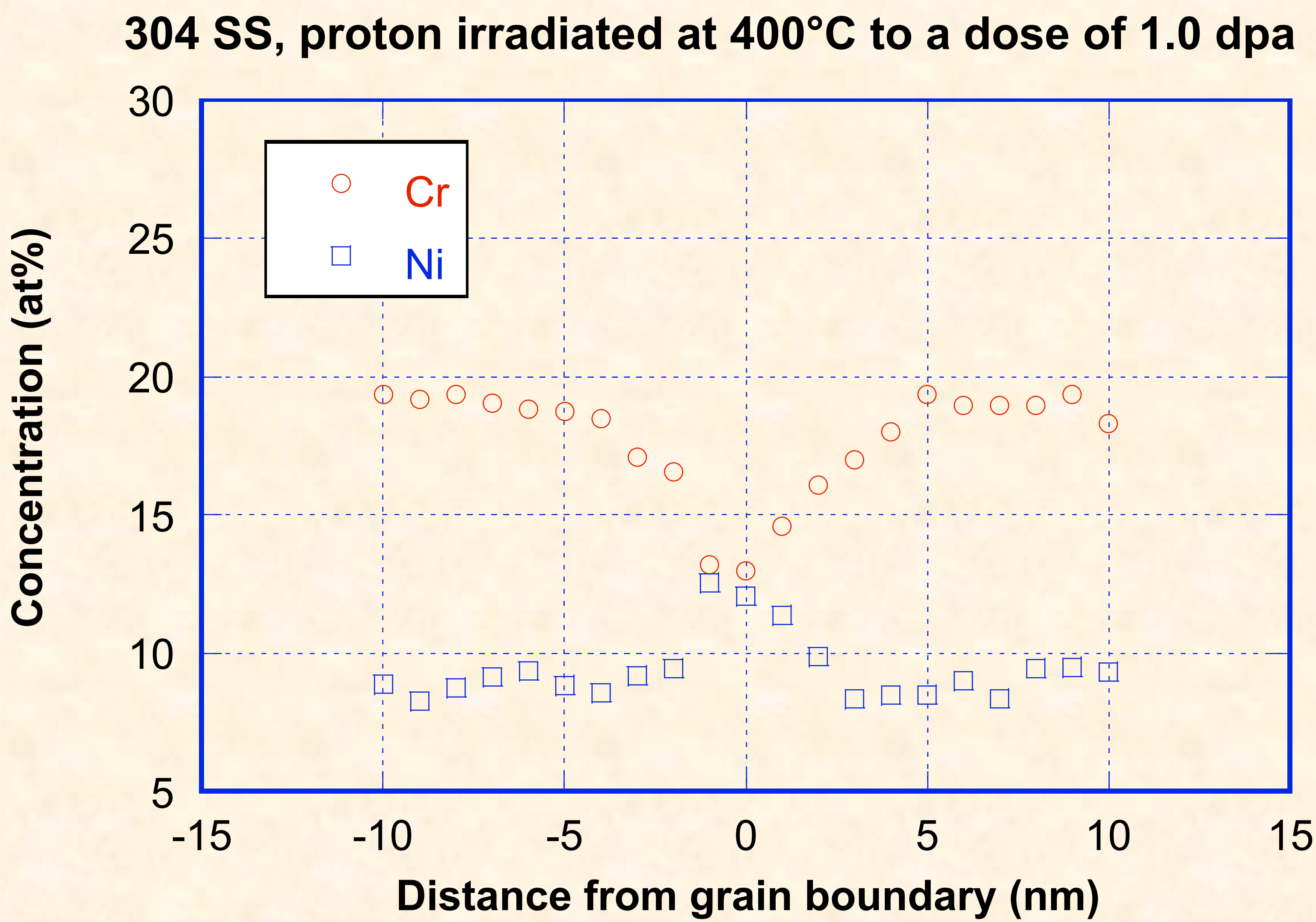
Diffusion-related forms of radiation-induced degradation

Radiation-Induced Segregation



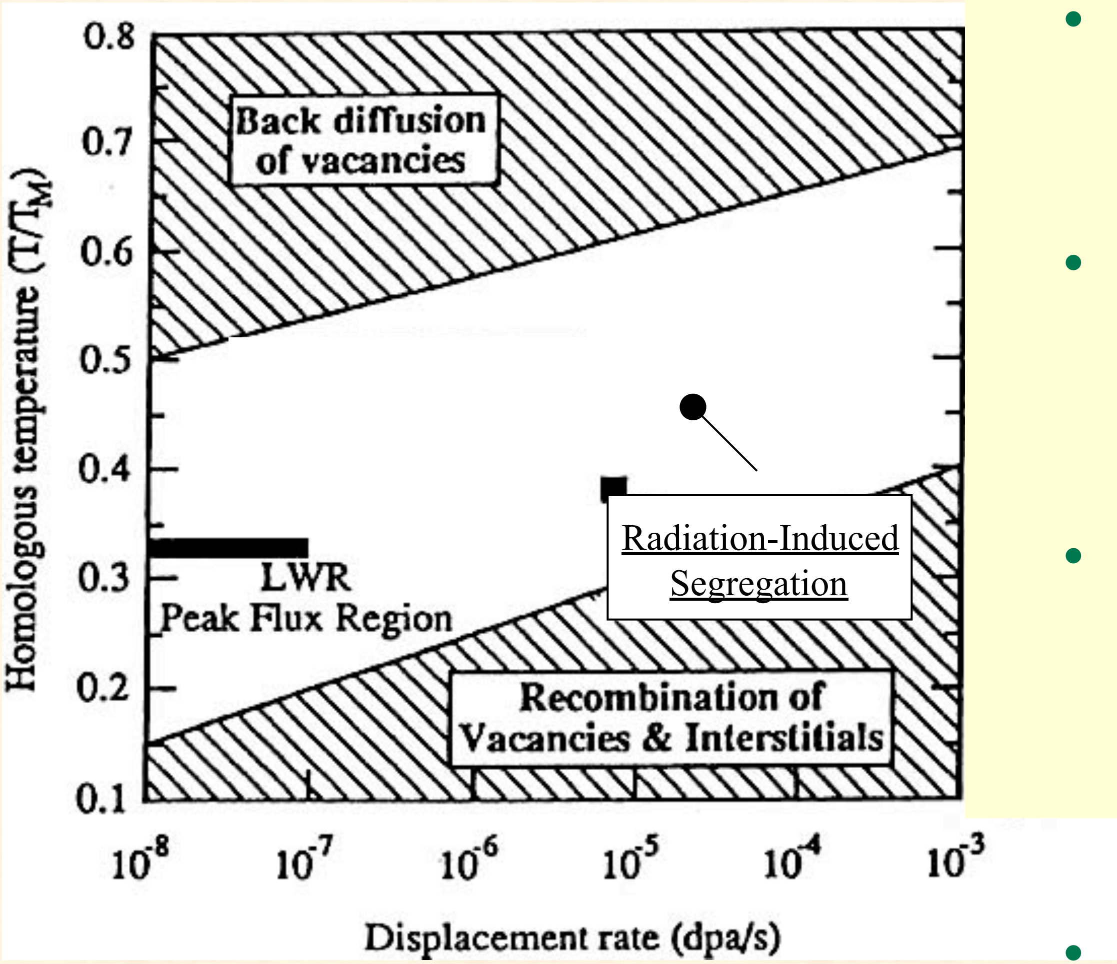
- **High concentrations of radiation-induced defects will migrate to defect sinks.**
- **Any preferential association between an atom and one type of defect will result in segregation.**

Grain Boundary Segregation



Source: T.R. Allen

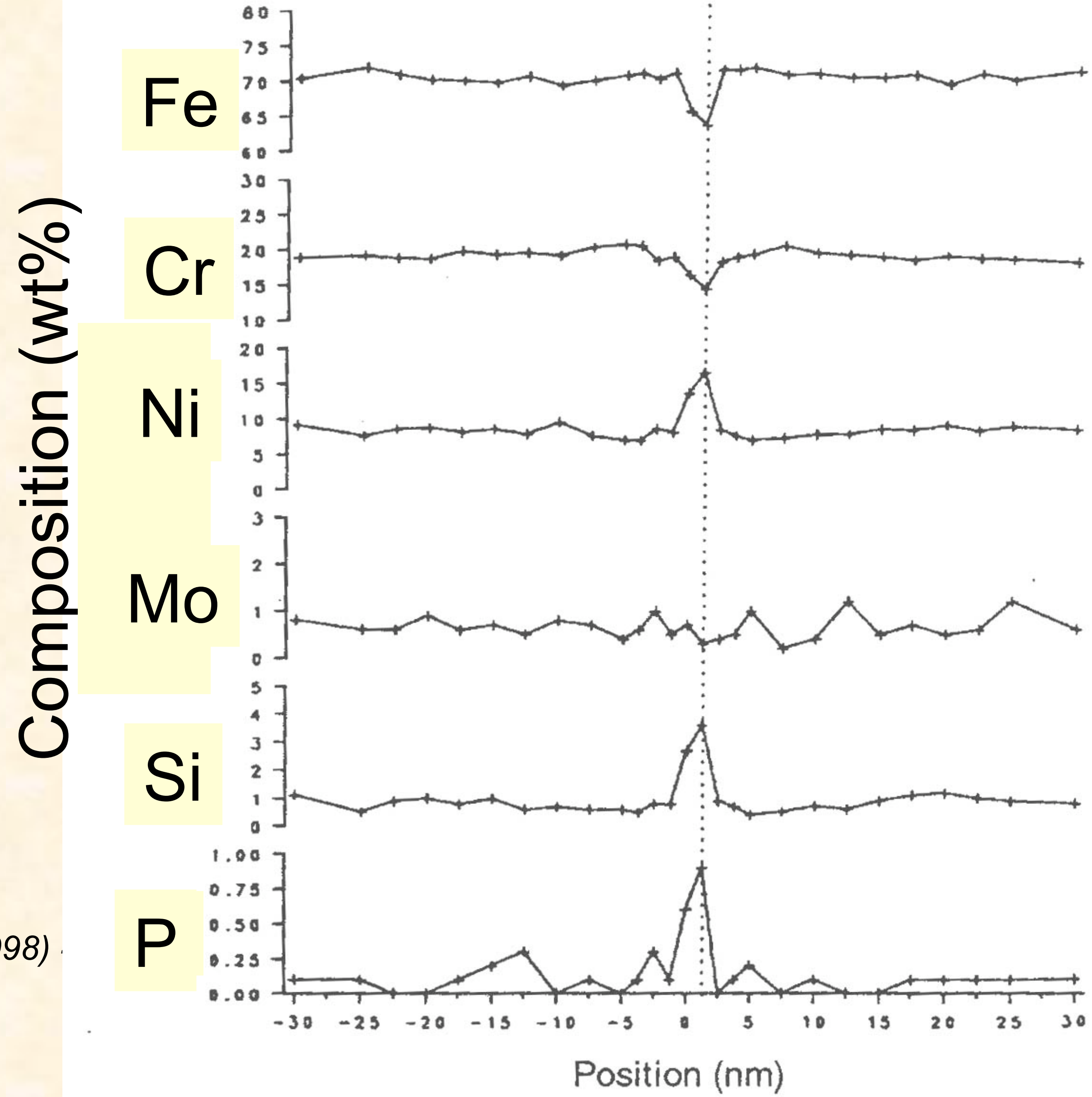
Radiation-induced segregation depends on temperature and dose rate



- The degree of segregation under irradiation will vary with temperature and dose rate.
- At low temperatures where defect mobility is limited, defect recombination dominates and RIS is minimal.
- At high temperatures where defect mobility and thermally induced defect populations are high, diffusion works to prevent or remove any composition gradients.
- At intermediate temperatures, however, RIS will occur.

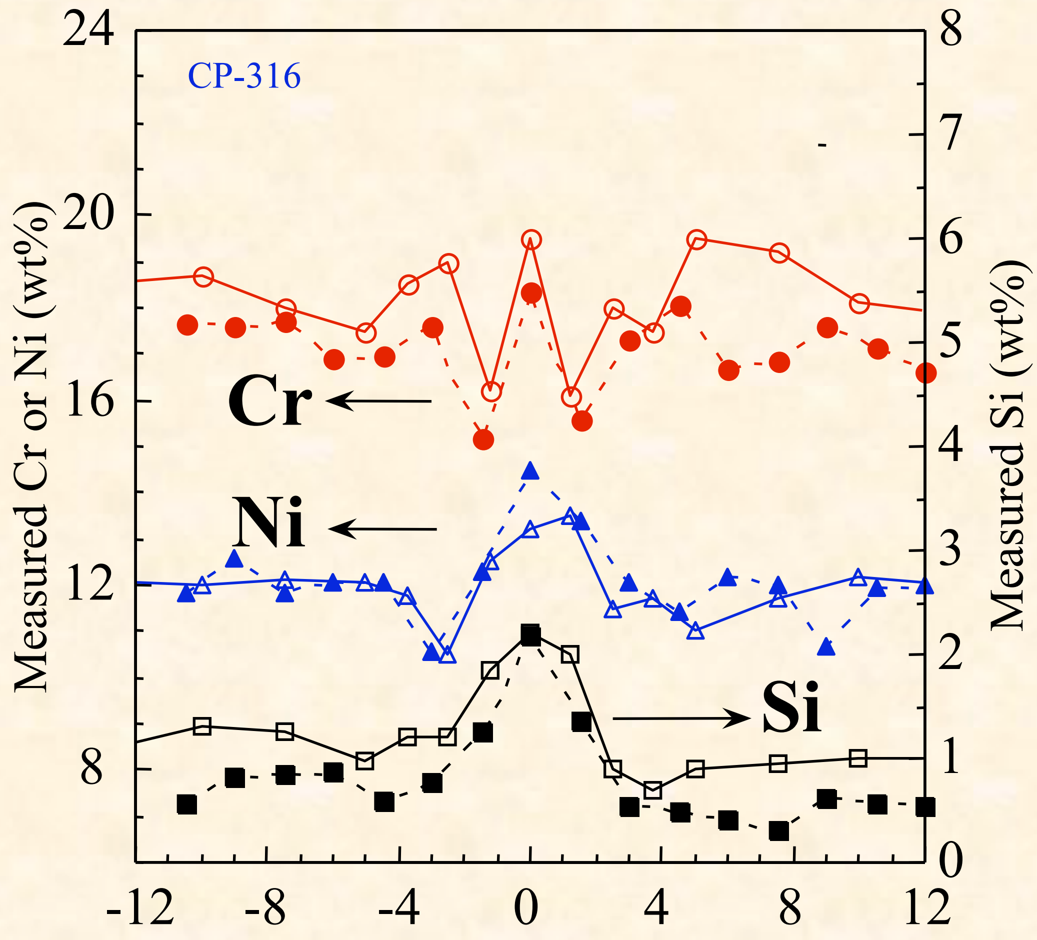
Concentration profiles for CP 304 SS after ~ 1022 n/cm2 at 288°C

Cr, Mo deplete
Ni, Si, P enrich
Fe depends on alloy composition

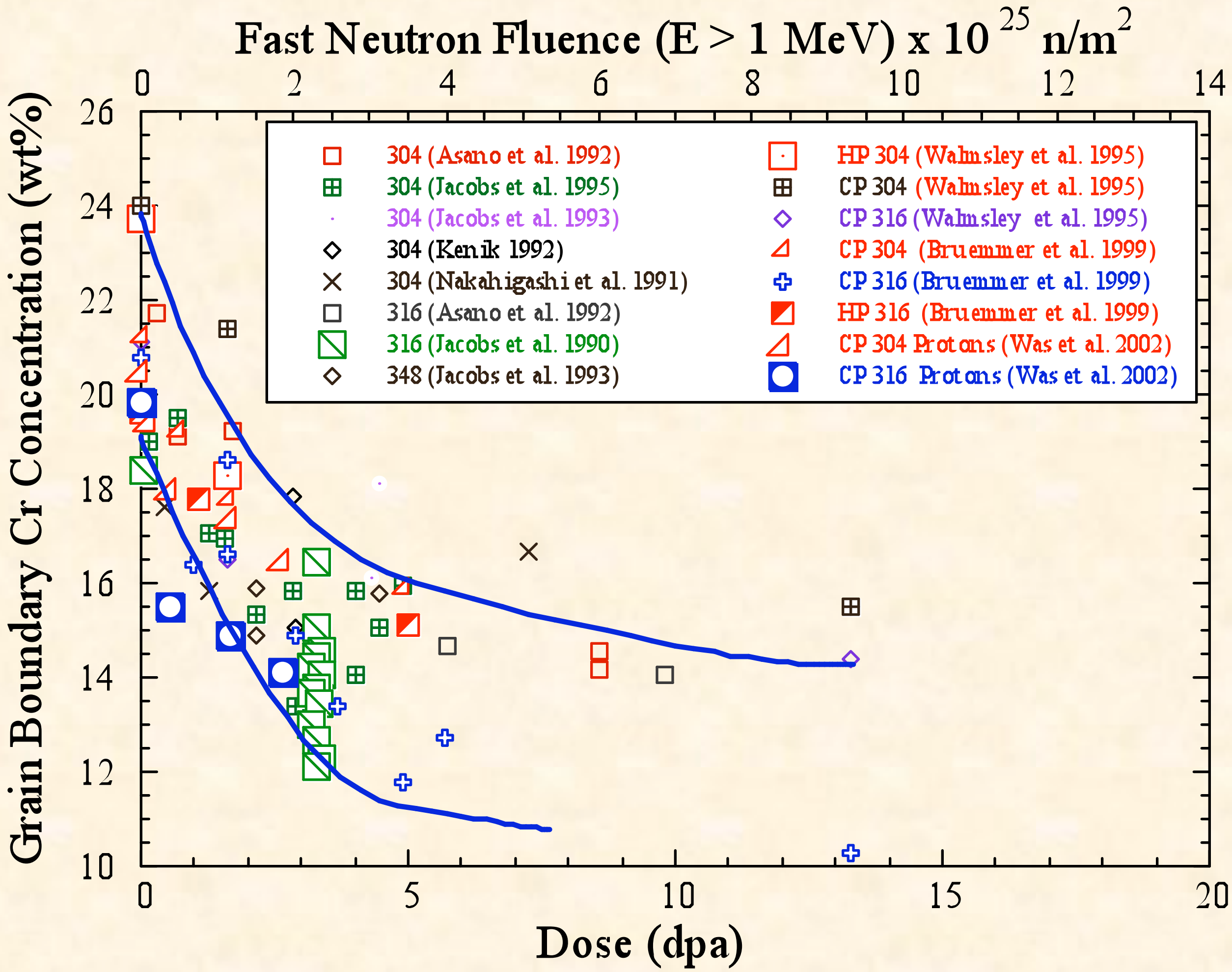


A. Jenssen et al, Corrosion 54 1 (1998)

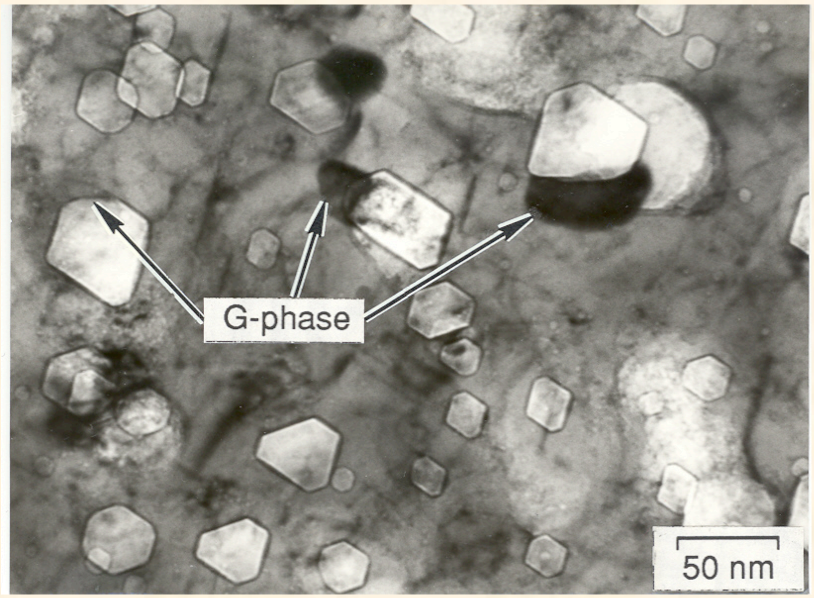
RIS can be quite complicated



Grain Boundary Chromium Depletion can be significant

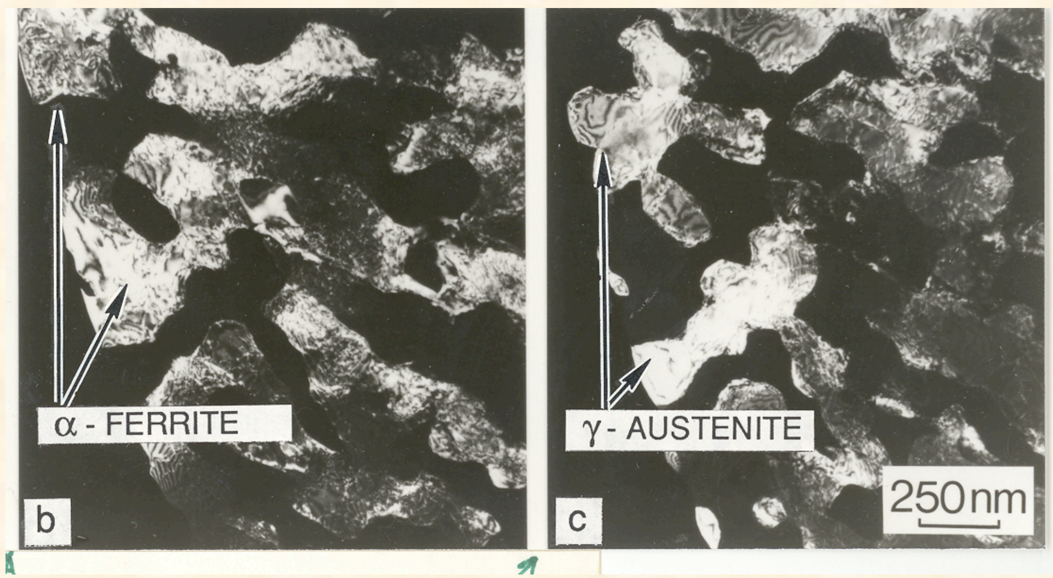


Radiation-Induced Segregation Can Lead to Precipitation

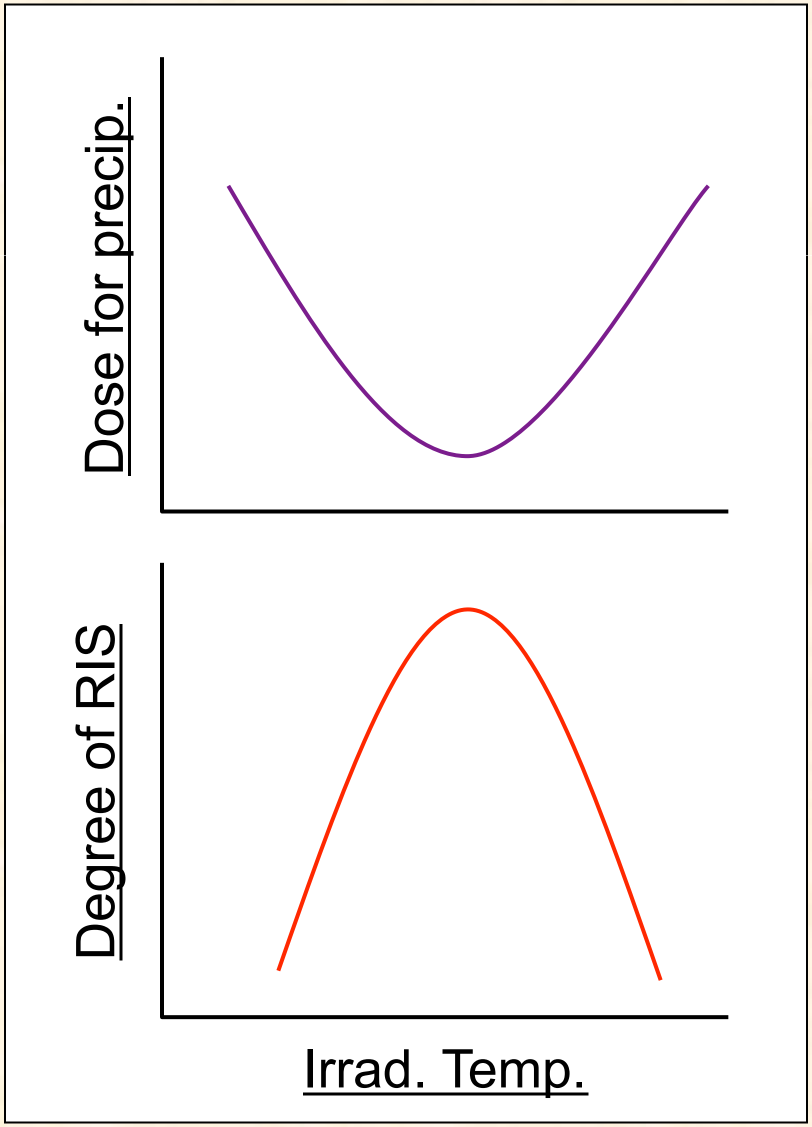


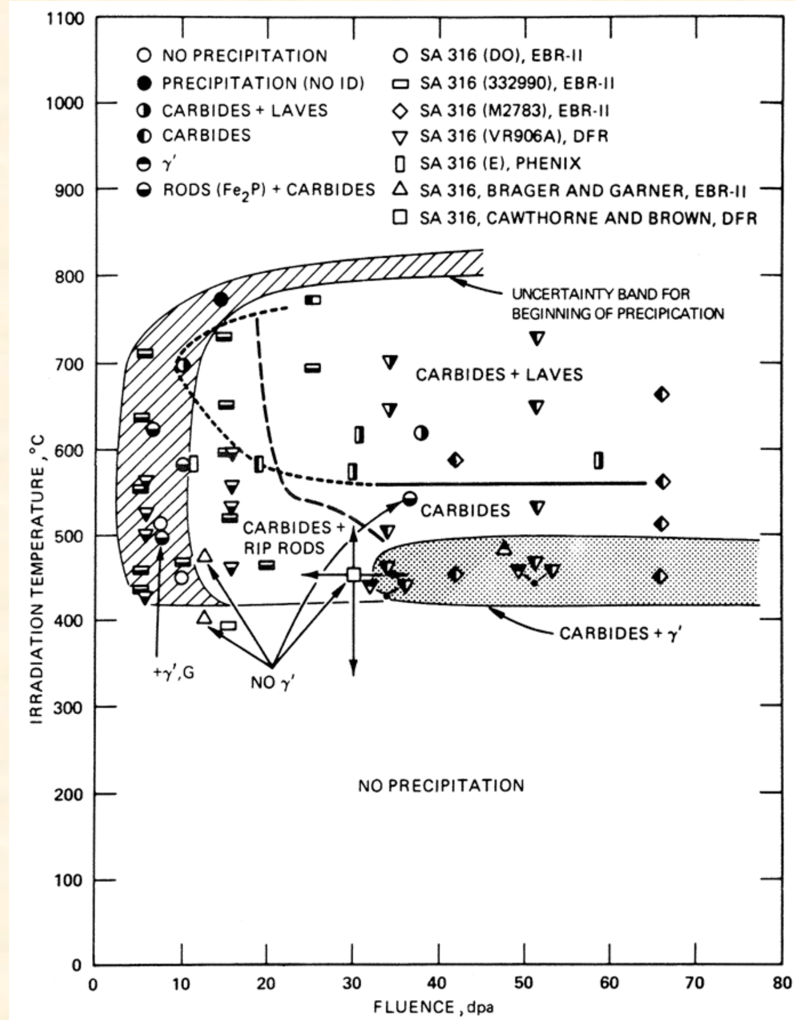
SA PCA steel irradiated at 500 C in ORR to 11 dpa (200 appm He), showing the association between radiation-induced G-phase silicide ($Mn_6Ni_{16}Si_7$) particles and the largest voids

- Microstructure of SA Fe-12Cr-15Ni-1Si austenitic stainless alloy irradiated in EBR-II at 575°C (<20 dpa), with TEM showing
- RIS causes the initially homogeneous γ-austenite matrix to decompose into Fe-Cr-Si ferrite and Fe-Ni austenite.



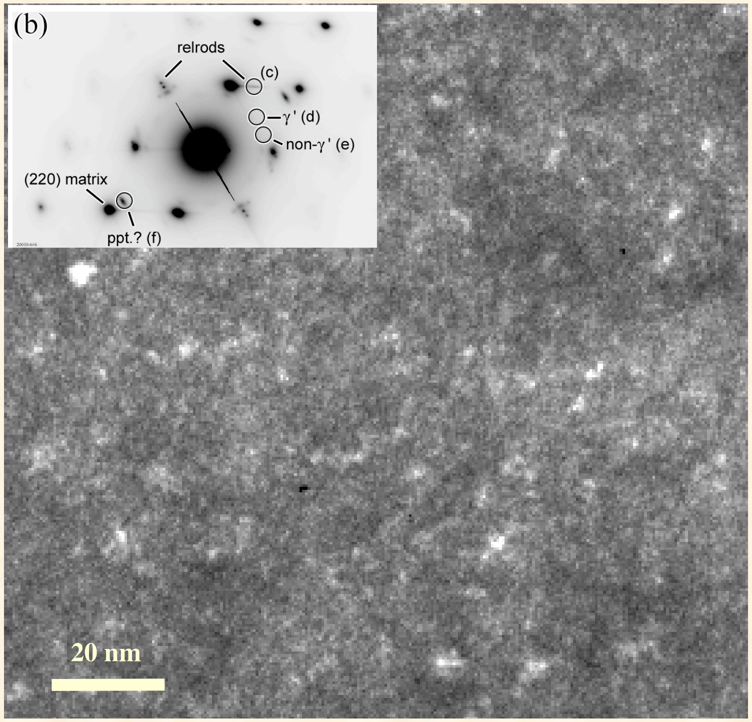
RIS and RIP will be very sensitive to the operating conditions



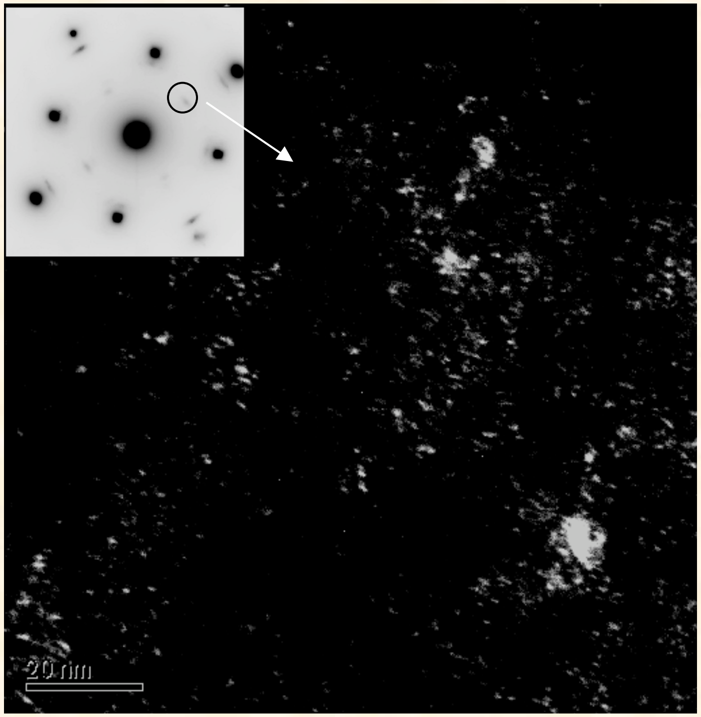


RIS and RIP have been widely observed in 316 SS alloys, and occurs in many other alloy systems.

Comparison of γ' in proton- and neutron-irradiated SS



Tihange baffle bolt:
neutron-irradiated to ~7 dpa
at 299°C*.



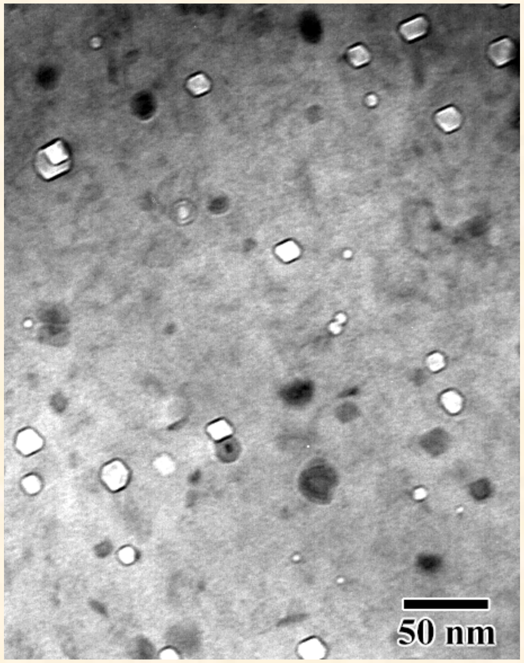
304+Si proton-irradiated to
5.5 dpa at 360°C.

•ATEM Characterization of Stress-Corrosion Cracks in LWR-Irradiated Austenitic Stainless Steel Core Components, PNNL EPRI Report, 11/2001.
•Image resized for equivalent scale.

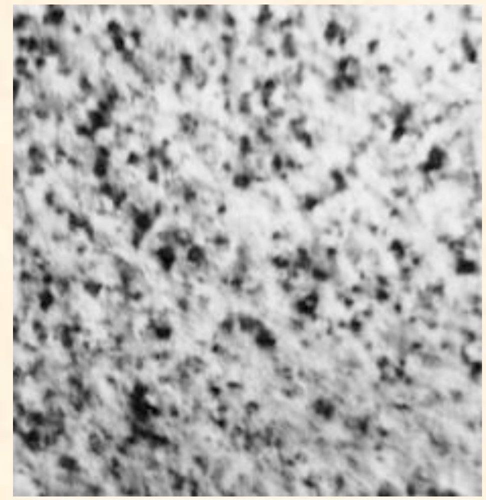
Fundamentals of Radiation Damage

Clustering-related forms of radiation-induced degradation

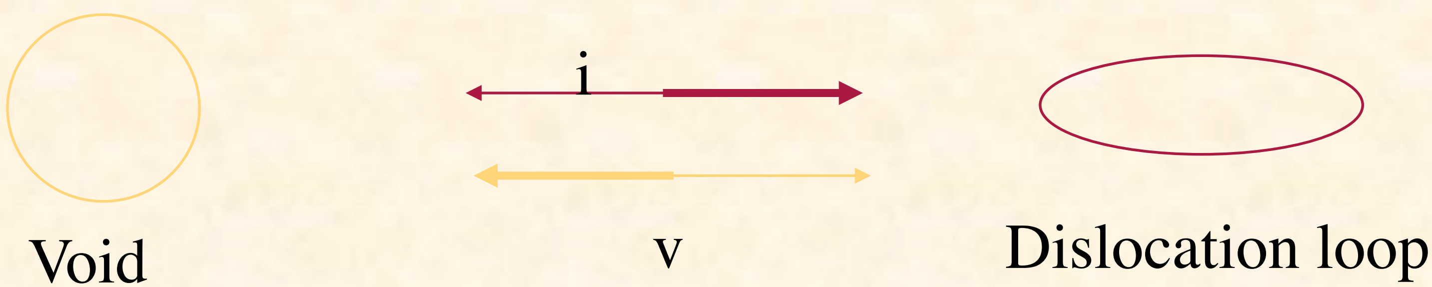
Clusters: voids and dislocation loops



- Process
  - Radiation produces point defects
  - Interstitials migrate preferentially to dislocations leaving excess vacancies to form voids
  - Both grow as they absorb more defects

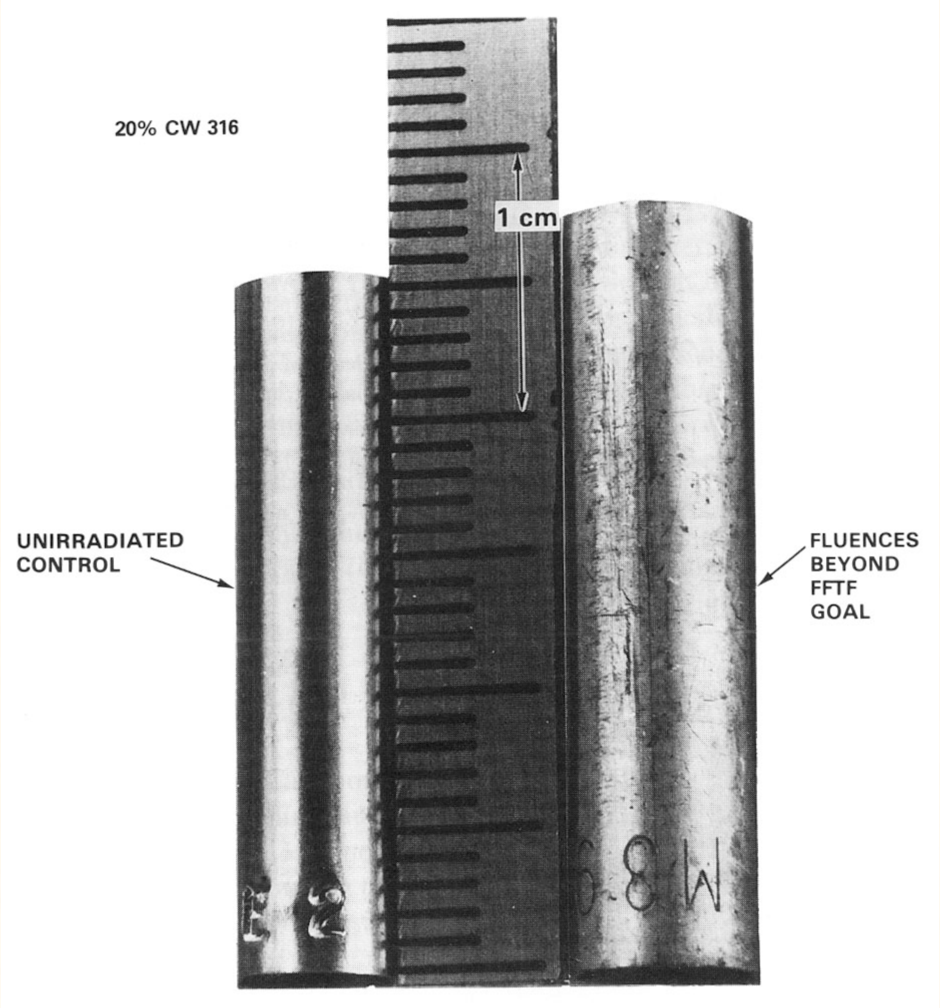


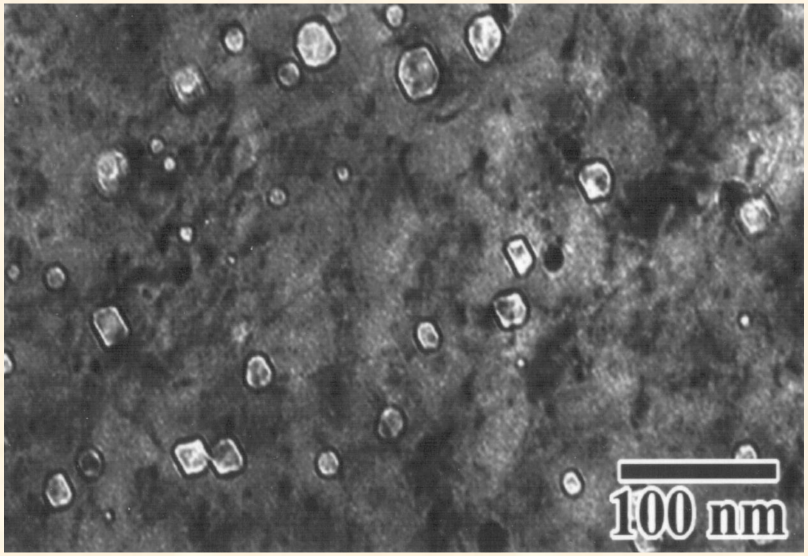
ρ_d = 17.0 x 10^{21} m^{-3}
d = 4.9 nm
50 nm



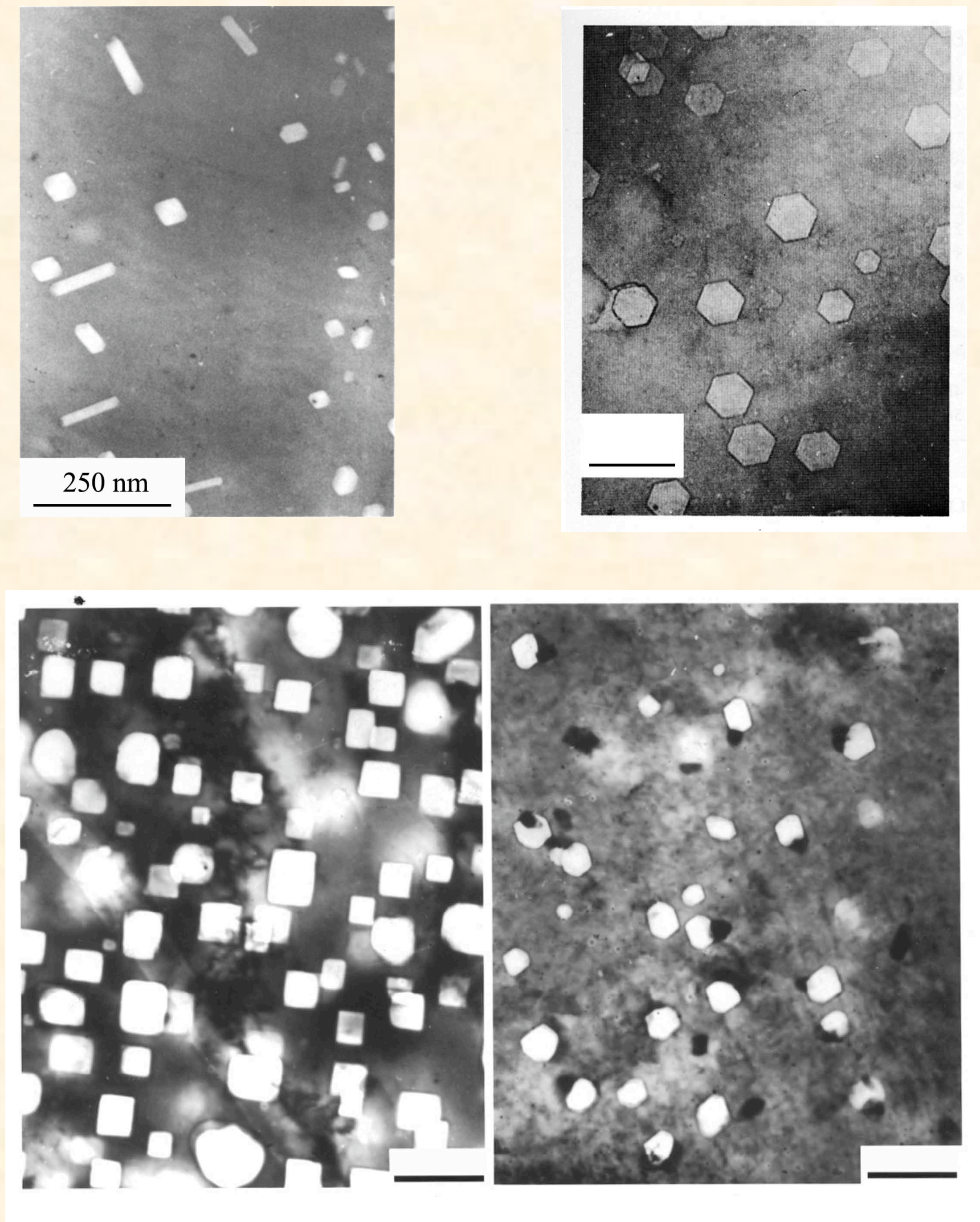
Source: T.R. Allen

Swelling is readily observed in many steels under various reactor conditions



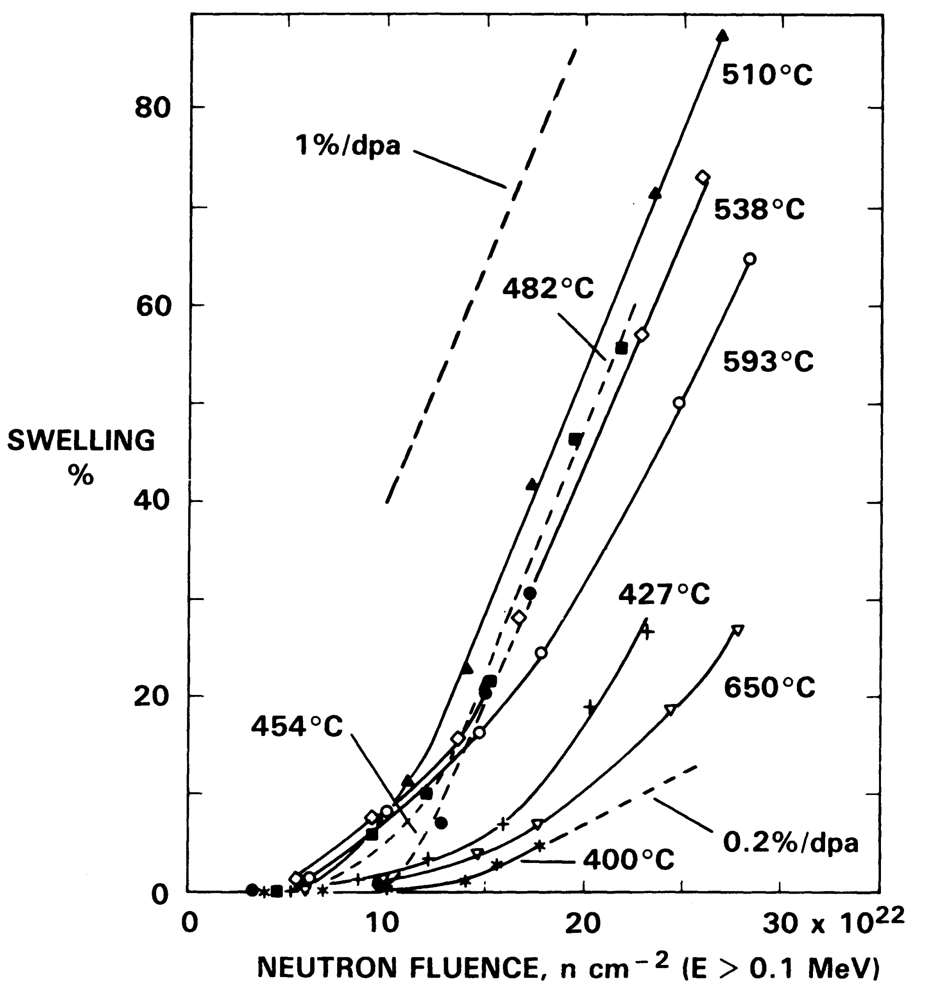


Source: F. Garner


250 nm

Swelling of 20% CW 316 stainless steel in EBR-II

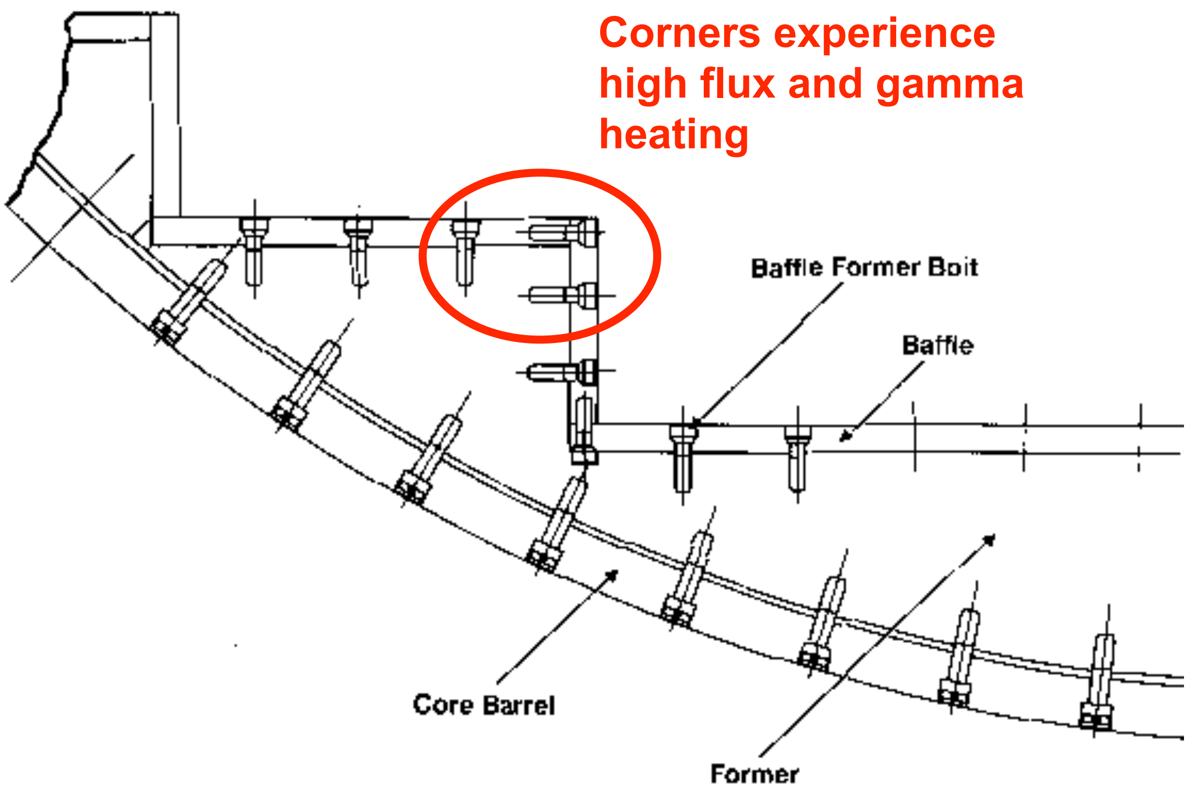
(Garner and Gelles, 1990)

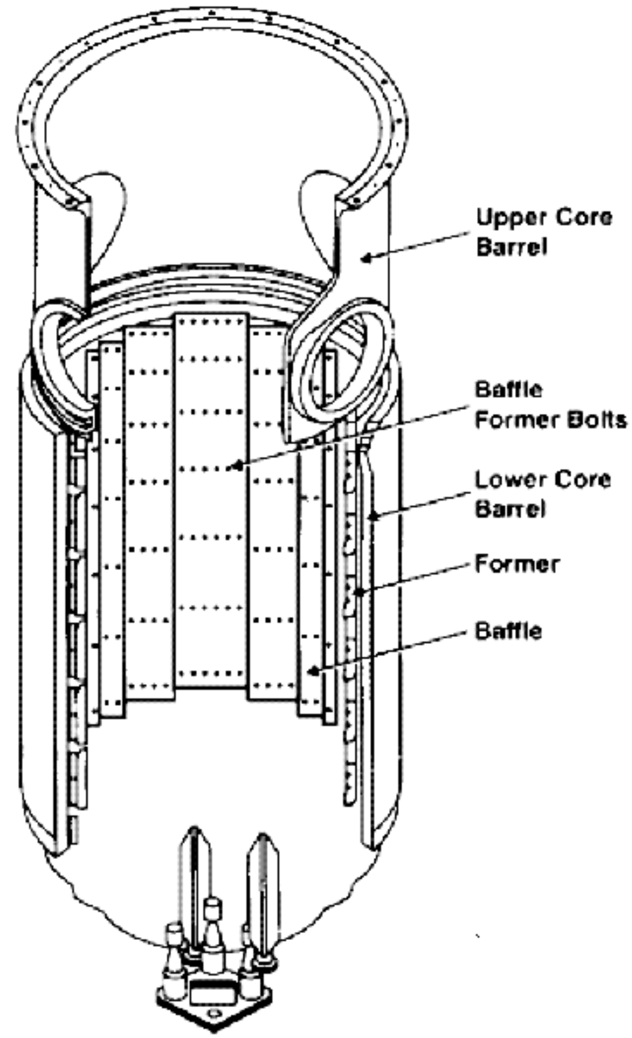


- Swelling occurs above temperatures above ~300ºC.
- The eventual swelling rate of 316 at all temperatures is ~1%/dpa.
- The temperature dependence lies only in the duration of the transient regime.
- The transient regime is very sensitive to any material or environmental variable.

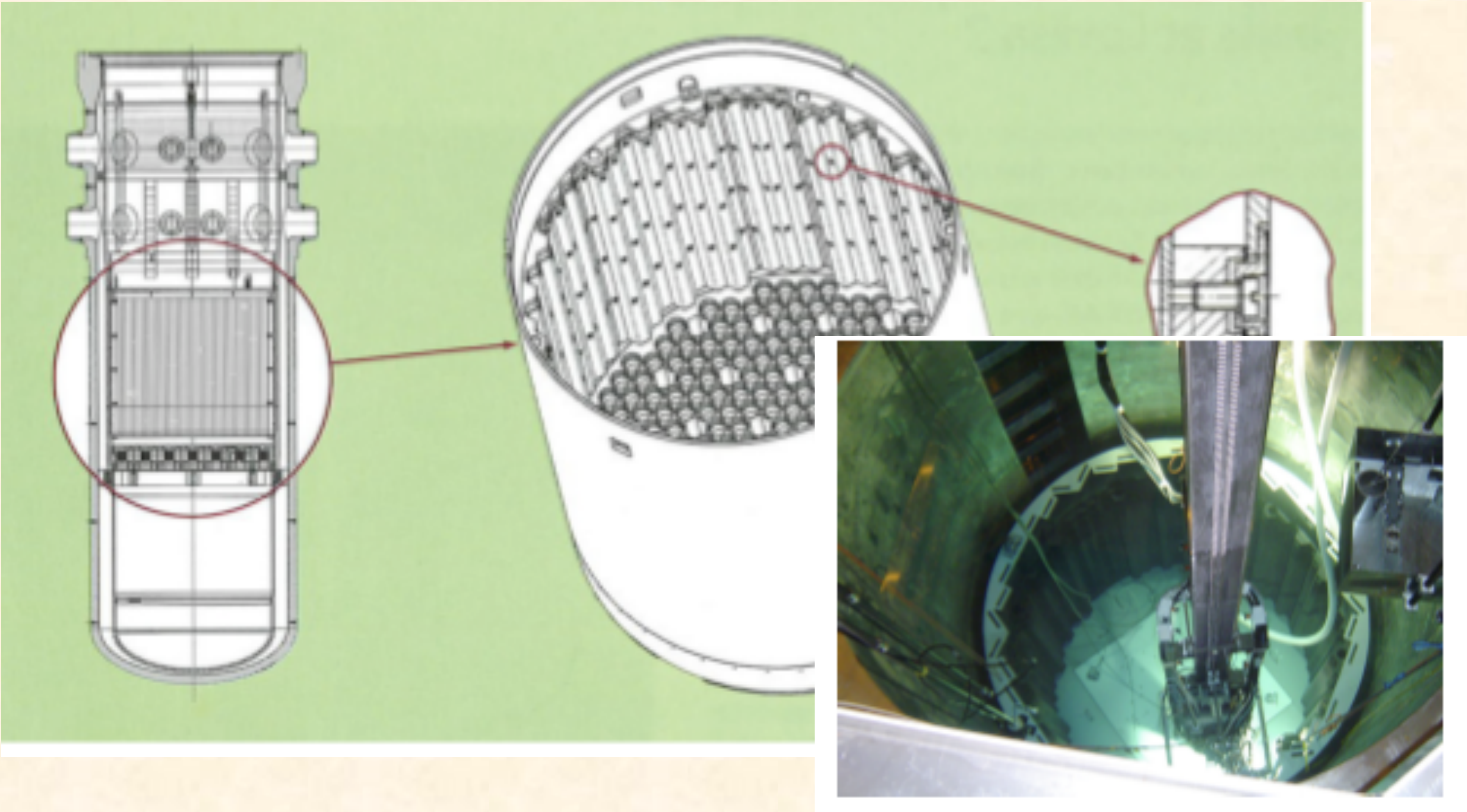
Source: F. Garner

Baffle bolts experience some of the highest fluences and temperatures in a PWR core



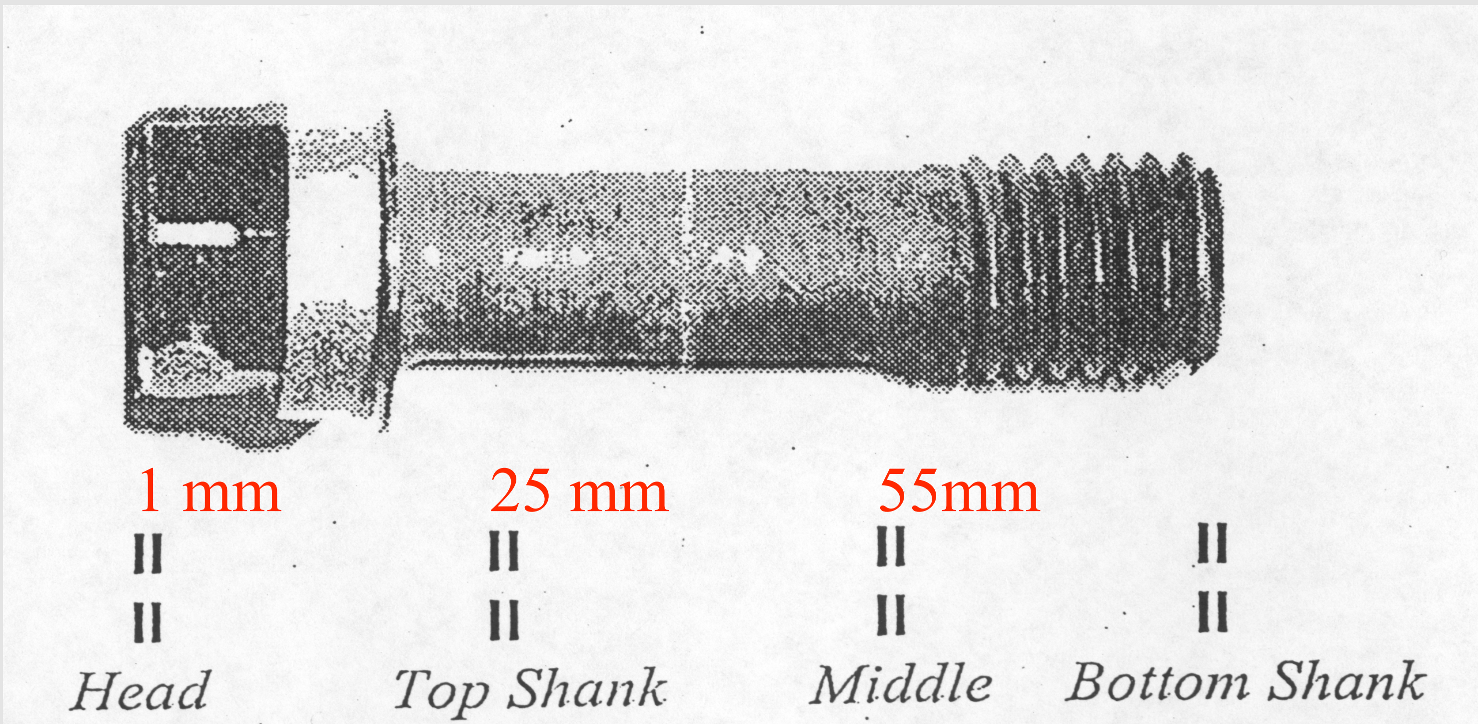


Baffle bolts experience some of the highest fluences and temperatures in a PWR core



Cutting diagram for 15% cold-worked 316 baffle bolt after 20 years in a PWR

Edwards, Garner, Oliver and Bruemmer, 2003



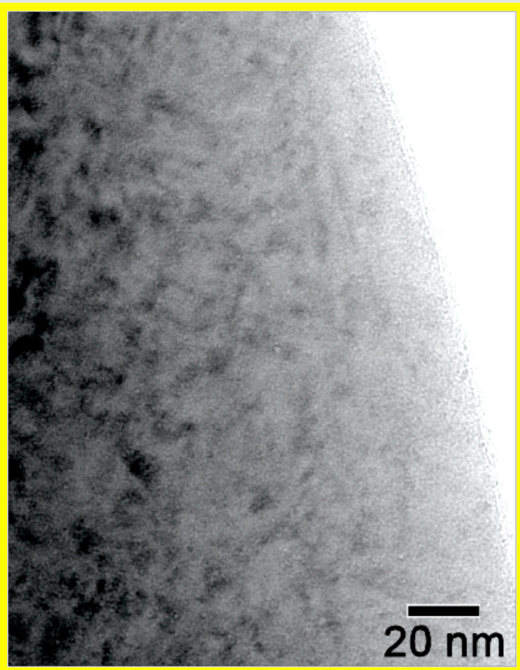
Head	Top Shank	Middle	Bottom Shank
19.5 dpa	12.5 dpa	7.5 dpa	
~320°C	~345°C	~330°C	~324ºC

Source: F. Garner

Swelling and helium content observed in CW 316SS baffle-former bolt

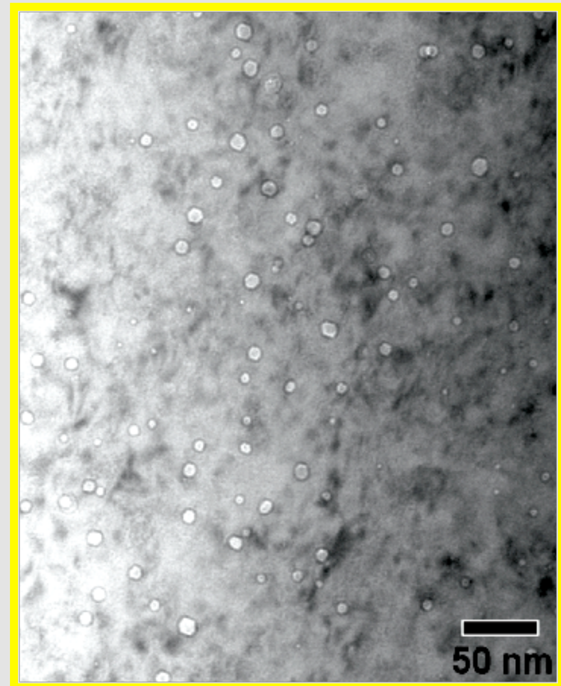
Edwards, Garner, Oliver and Bruemmer, 2003

<.0.01%, 71 appm Helium | Both at ~0.2% swelling , 53 and 49 appm Helium



Bolt Head, 1 mm
19.5 dpa, ~320°C

Top Shank, 25 mm
12.5 dpa, ~343°C

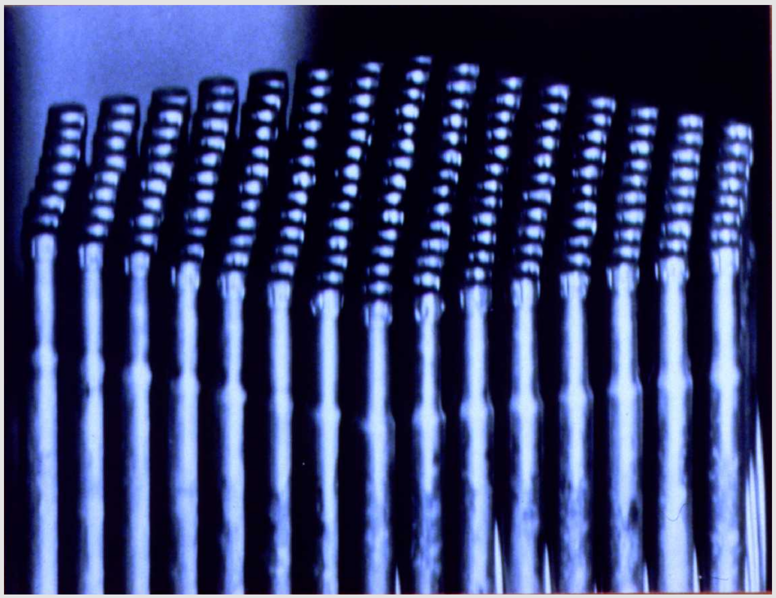


Near Threads, 55 mm
7 .5 dpa, ~333°C

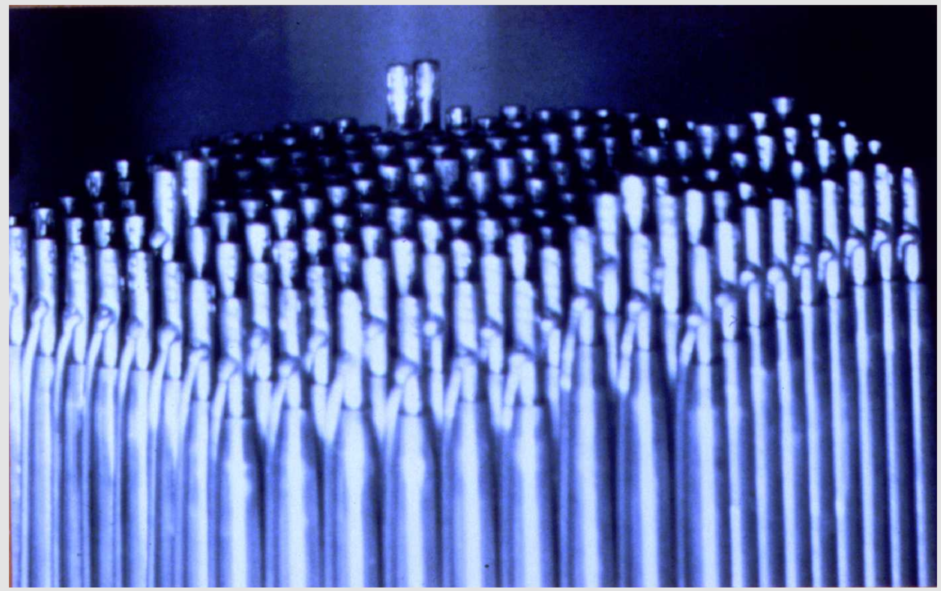
Source: F. Garner

Swelling can create tolerance problems

FFTF Fuel Pin Bundles



HT-9, no swelling

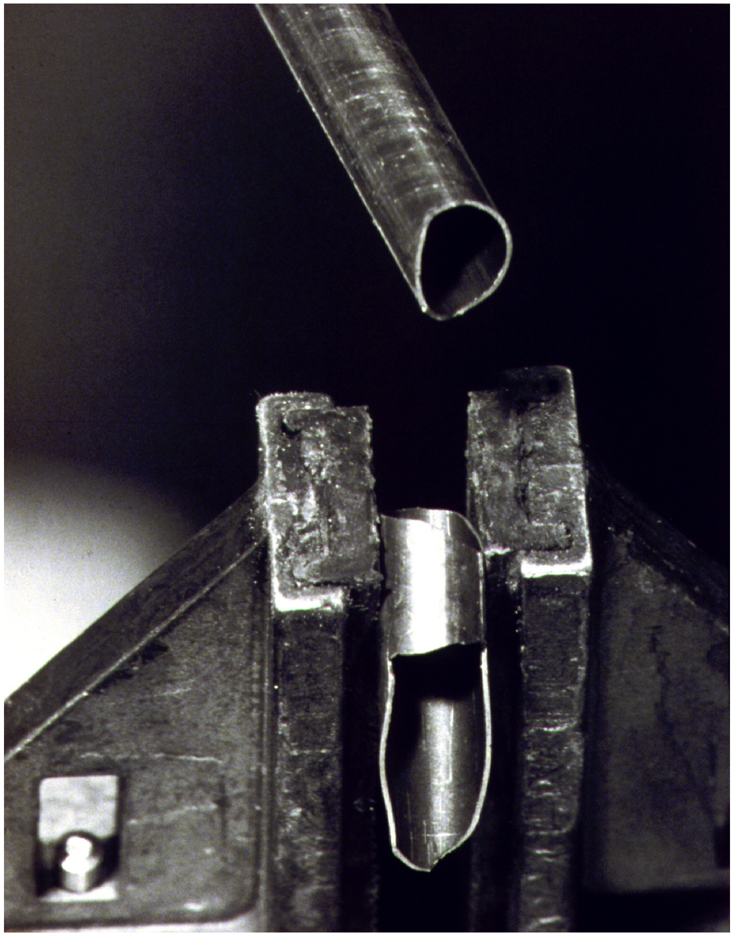


316-Ti stainless, swelling

Source: F. Garner

Severe void-induced embrittlement

DEPARTMENT OF Engineering Physics
College of Engineering University of Wisconsin-Madison

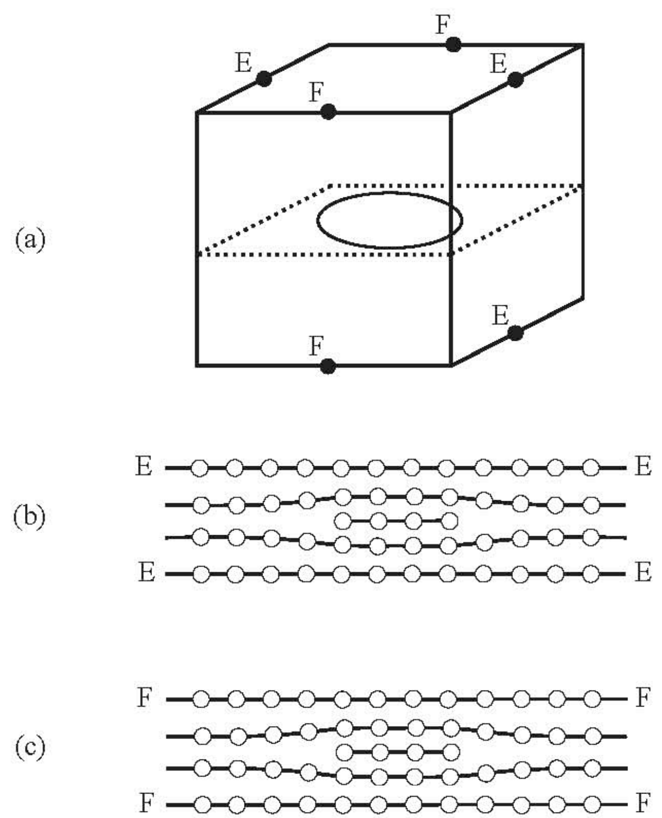


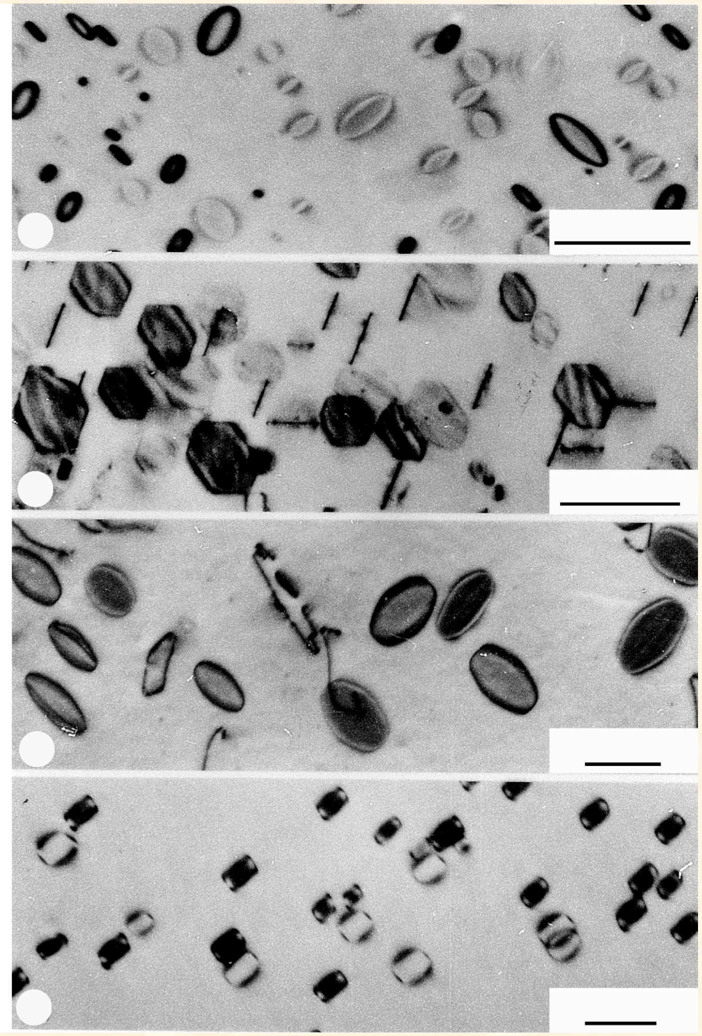
- 14% swelling
- Failure occurred during clamping in a vise at room temperature.
- Tearing modulus has fallen to zero, with no resistance to crack propagation.
- 316 stainless steel irradiated at ~400ºC
- Embrittlement threshold at ≲10% swelling.

Source: F. Garner Source: T.R. Allen

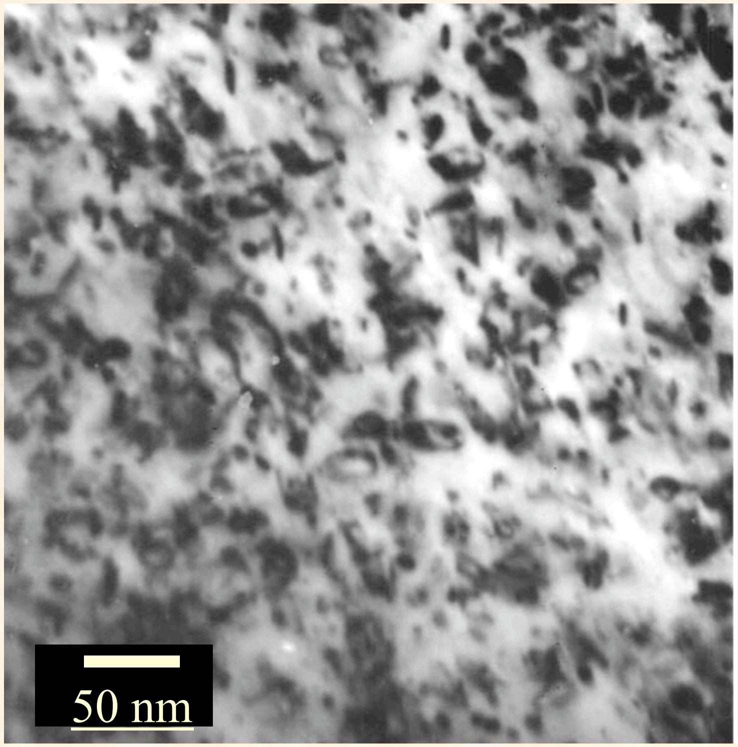
Interstitials (or vacancies) can also cluster into discs

Faulted (Frank) Loop

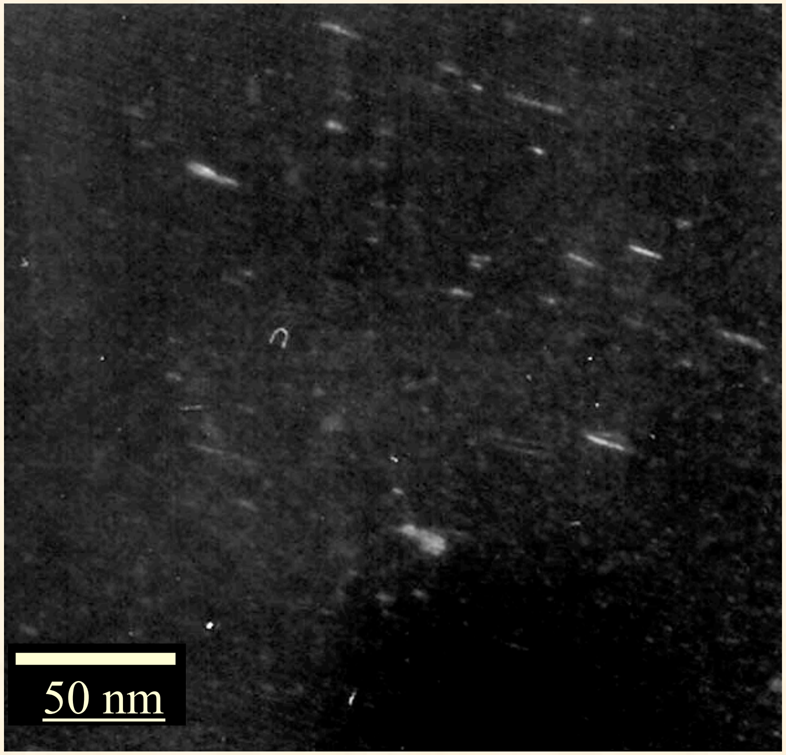




Dislocation loop microstructure is observed using transmission electron microscopy



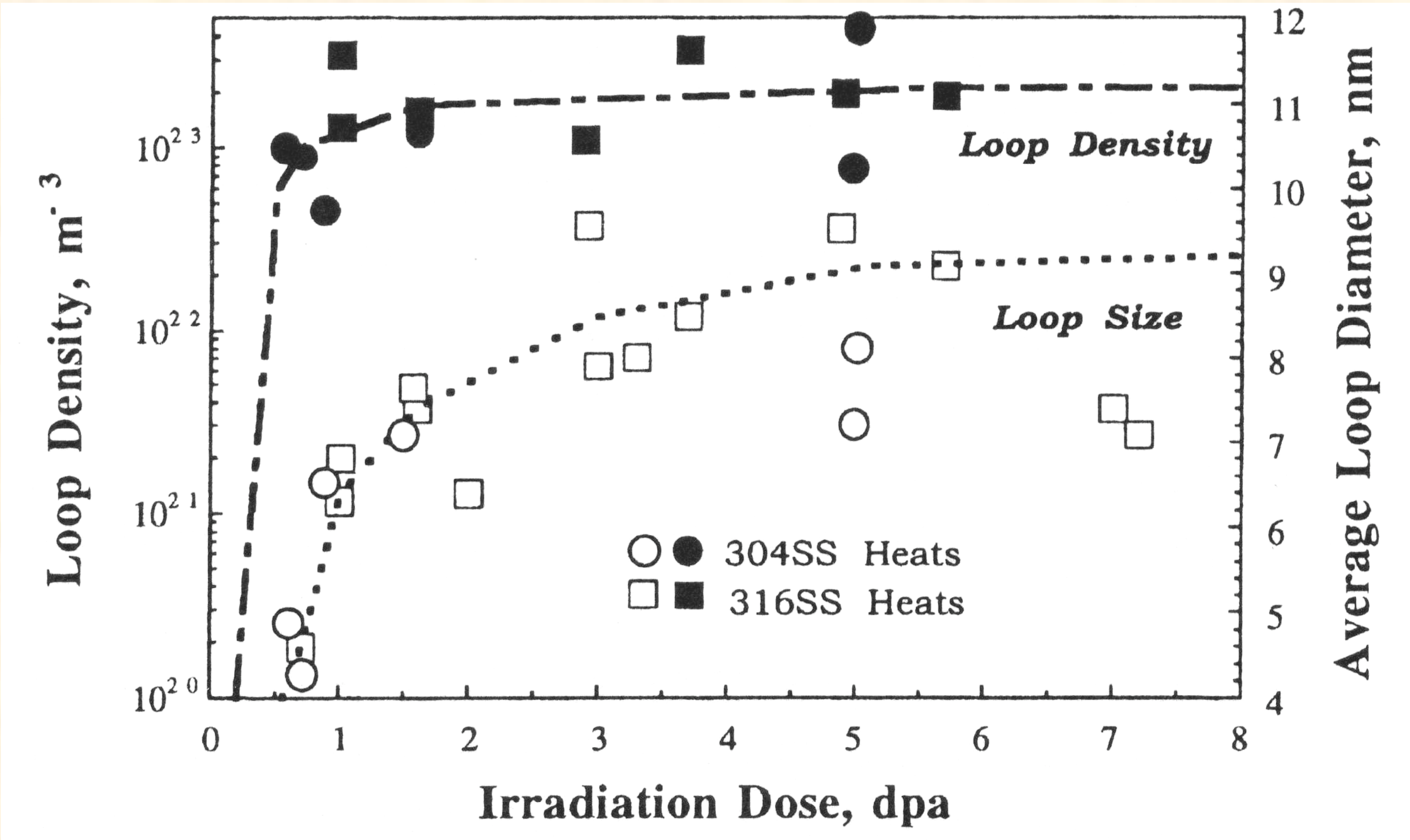
Bright Field



Dark Field

CP-304 SS irradiated to 0.55 dpa with protons at 360°C

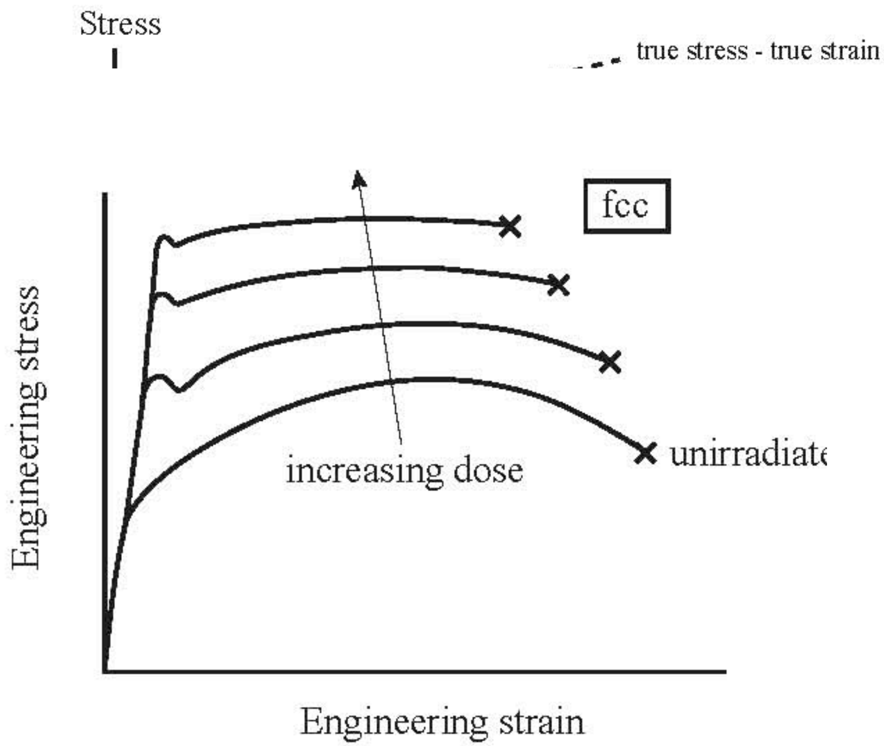
Behavior of Dislocation Loop Size and Number Density



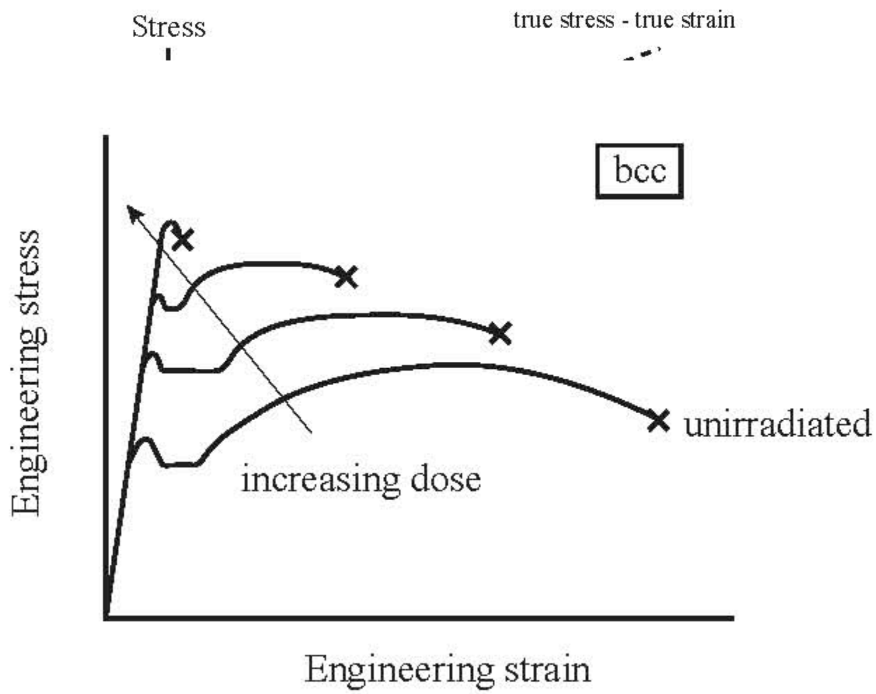
S. Bruemmer et al. JNM 274 (1999) 299.

Irradiation-induced defects and microstructure also influence stress-strain curves

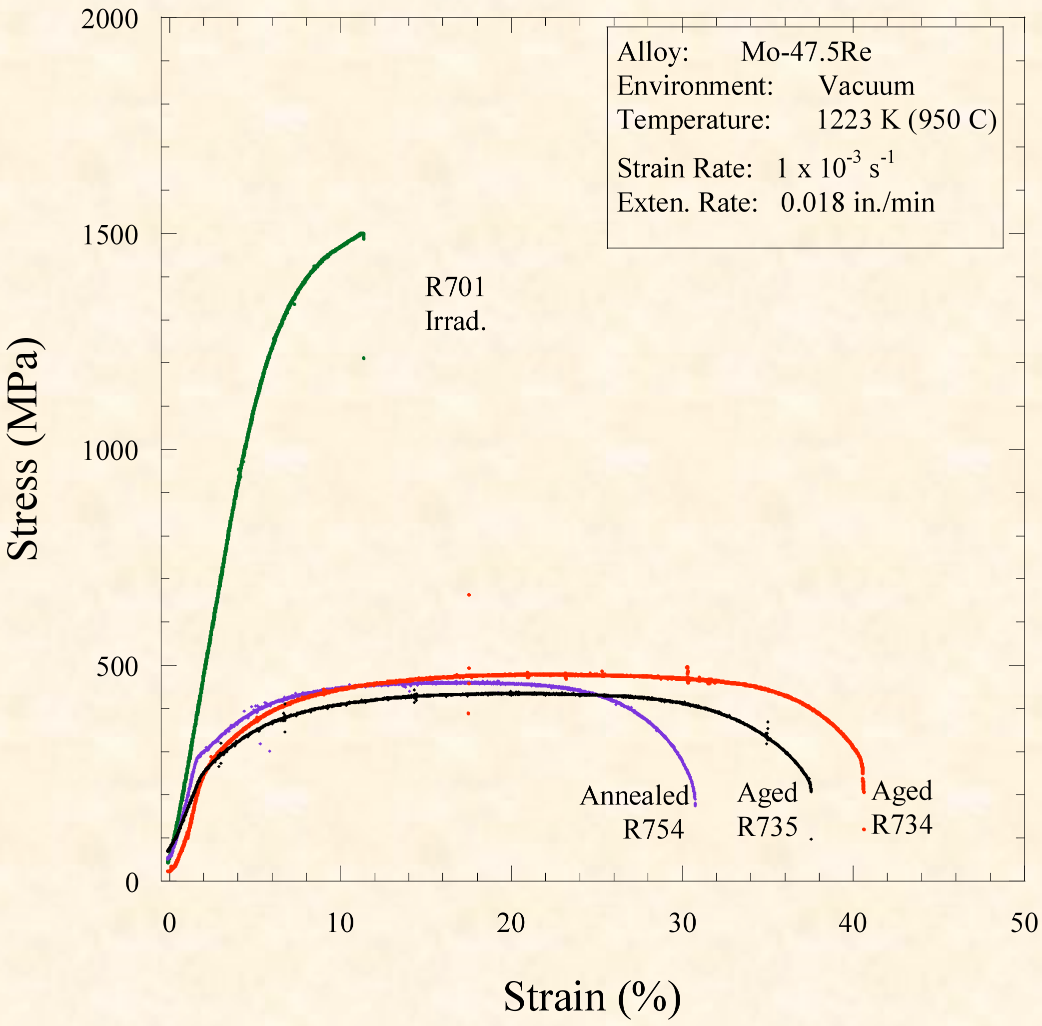
fcc



bcc

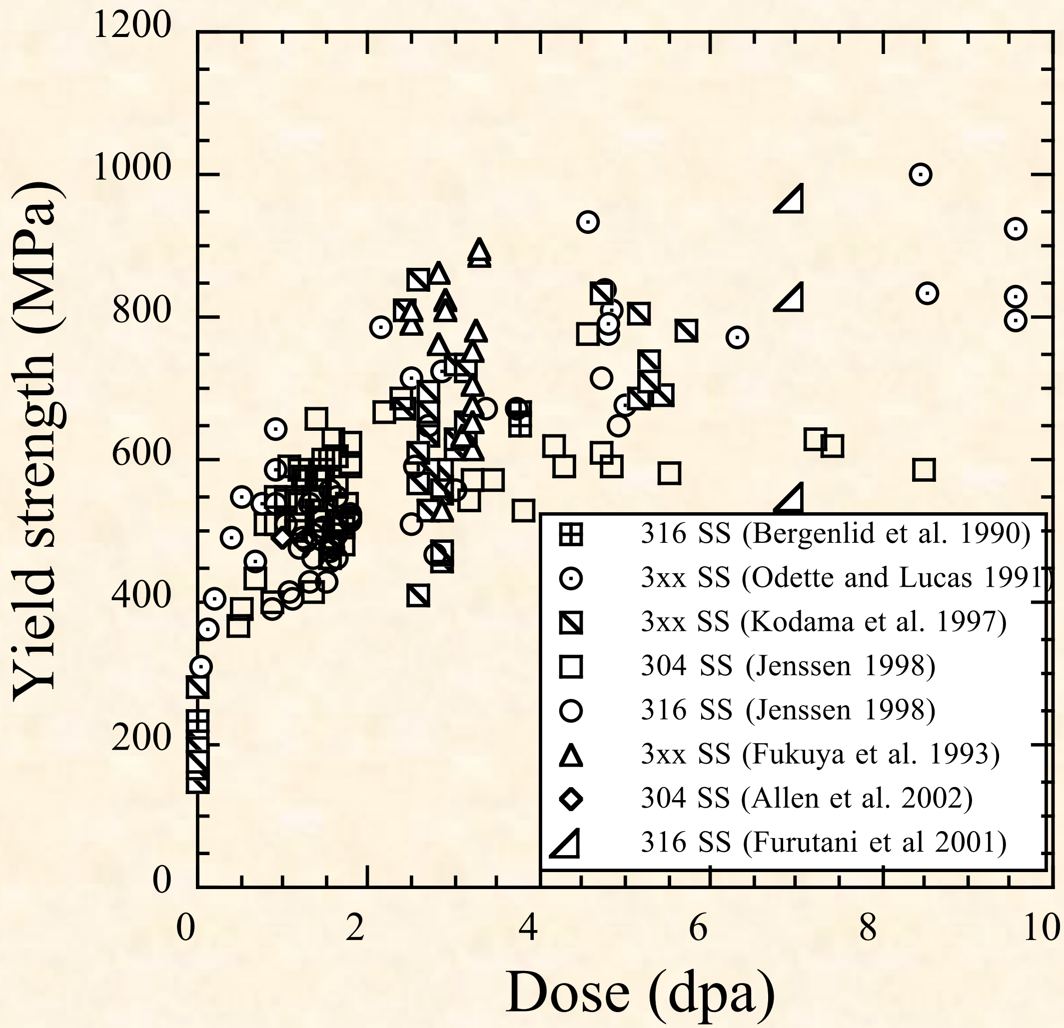


Radiation hardening is observed in many classes of materials

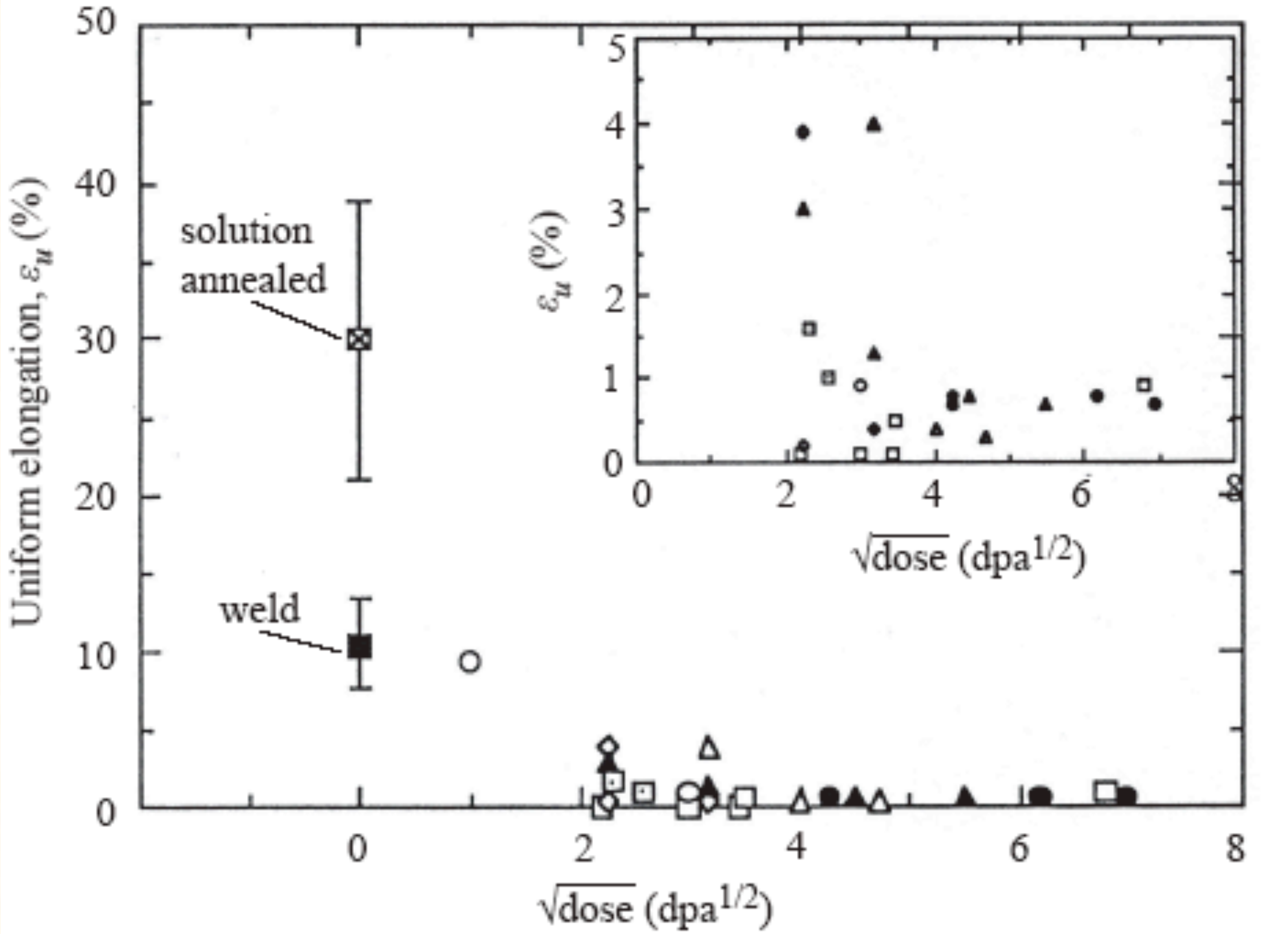


- **This hardening is also observed in many different materials.**
- **Increases in YS and UTS are commonly observed.**
- **Irradiation also results in a drop in ductility.**

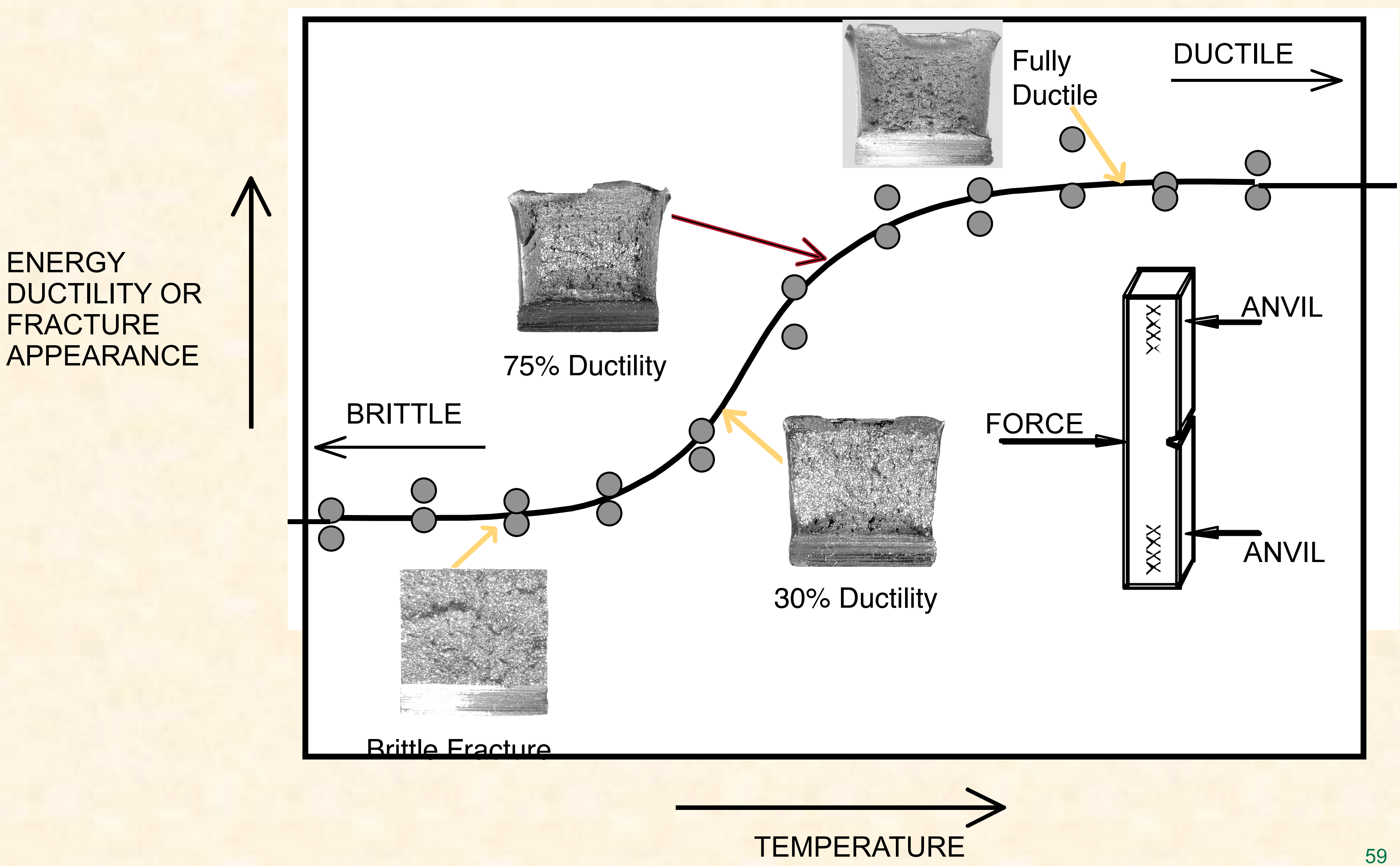
Irradiation hardening is widely studied in stainless steels



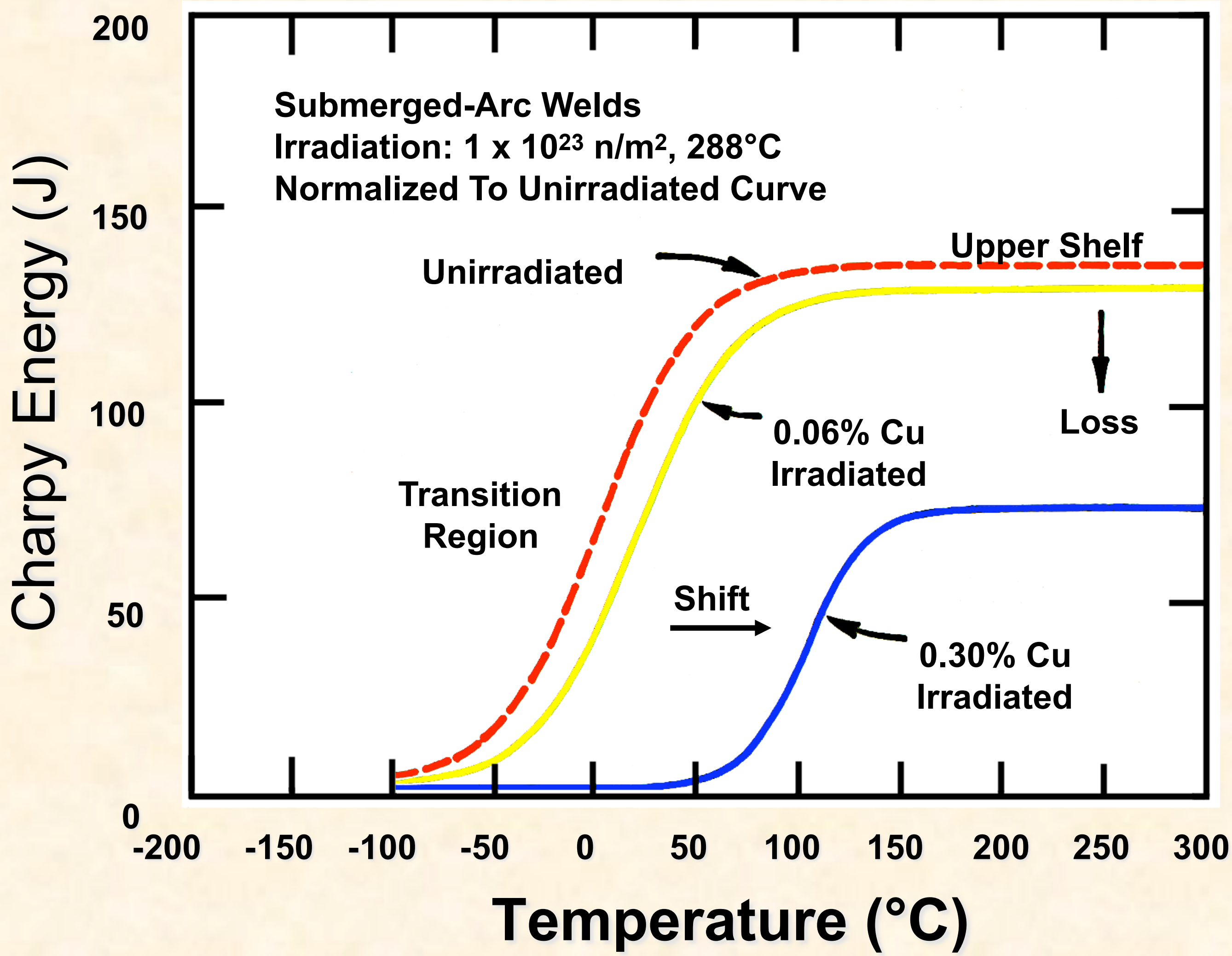
Hardening also results in a loss of ductility



Loss of ductility can lead to loss of toughness or even brittle failure

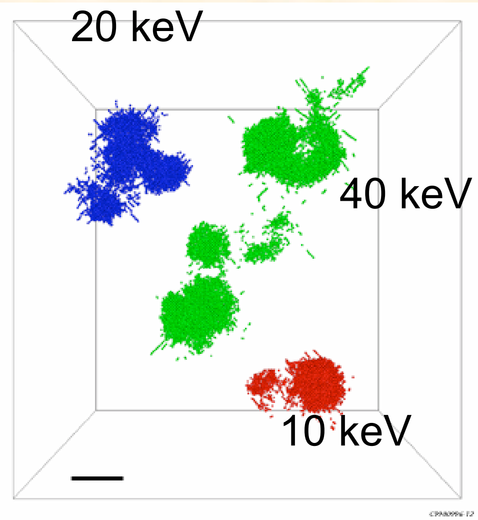


Radiation-embrittlement and the DBTT are a key concern in RPV steels

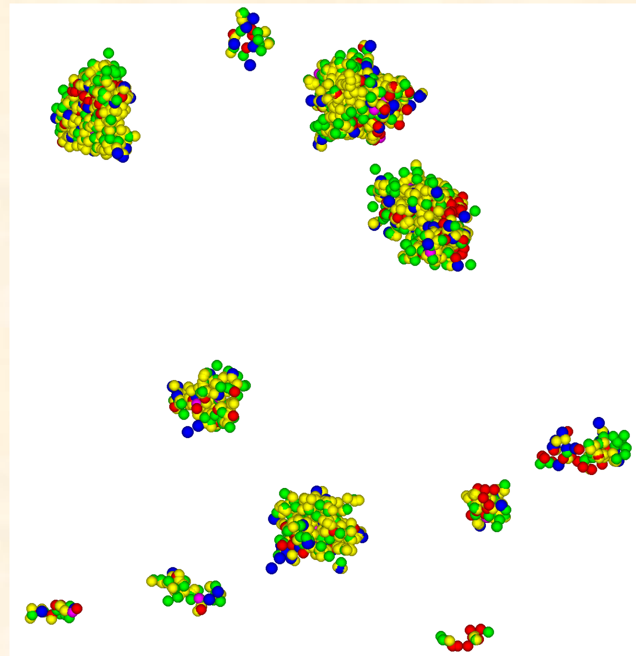


Irradiation Causes Ductile/Brittle Transition Temperature Shift and Upper Shelf Energy Loss — Copper Increases The Effect

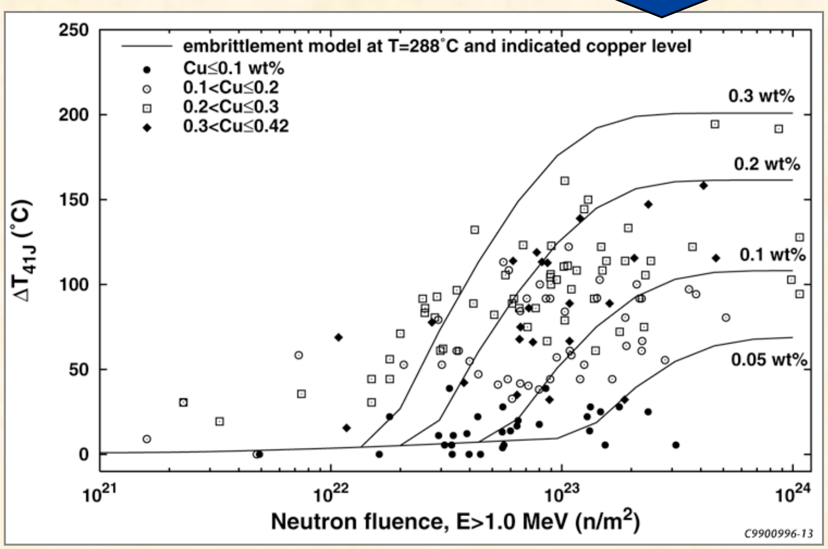
Microstructural Investigations Such As Atom Probe Tomography Are Used to Reveal Mechanisms of Irradiation Embrittlement



Neutron radiation produces an extremely high number density of nanoscale copper-, manganese-, nickel-, silicon-, and phosphorus-enriched precipitates.



Fe Cu Ni Mn Si P atoms

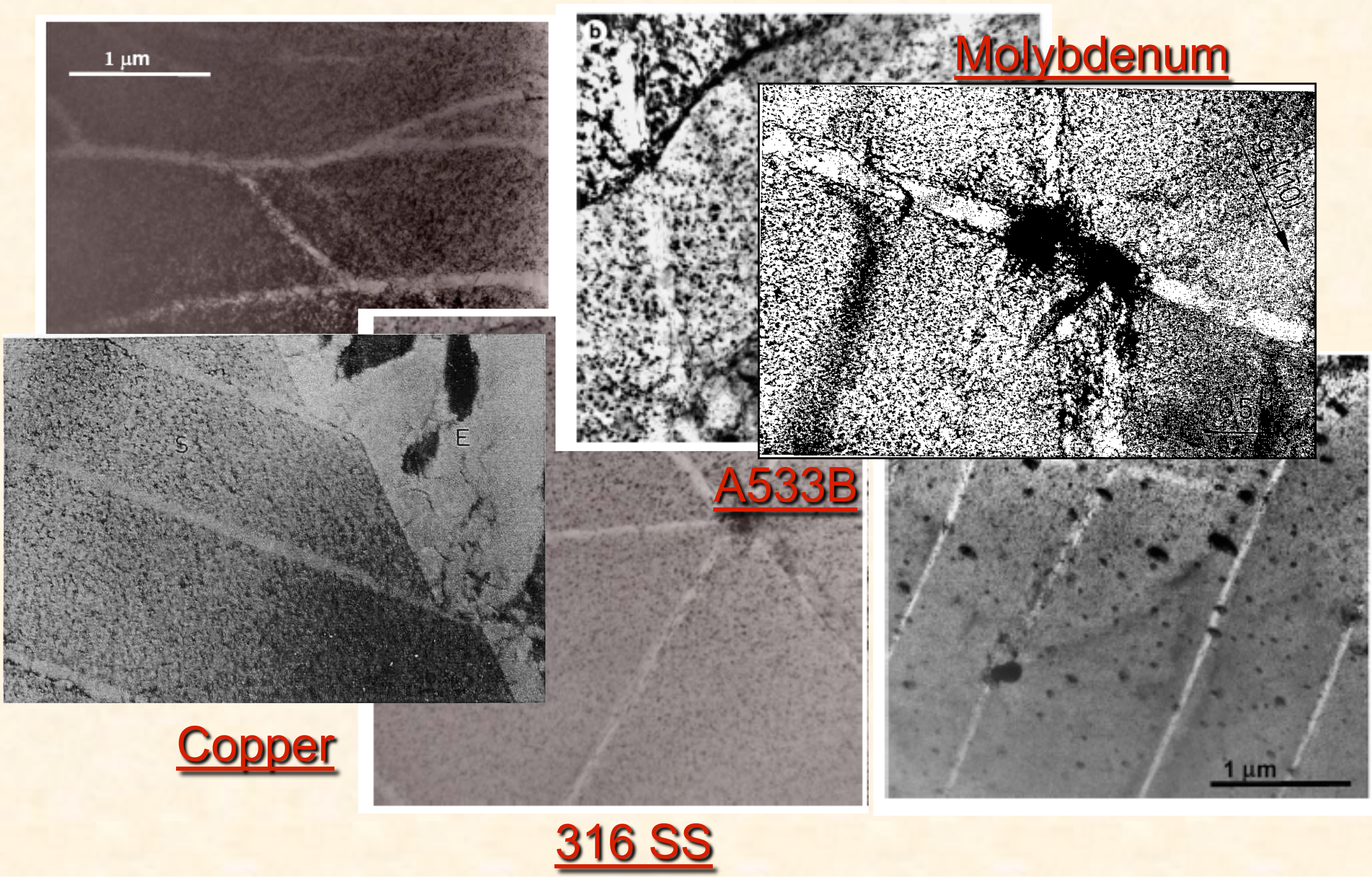


Combination of experimental, modeling, and microstructural studies leads to advances in predictive capability.

Fundamentals of Radiation Damage

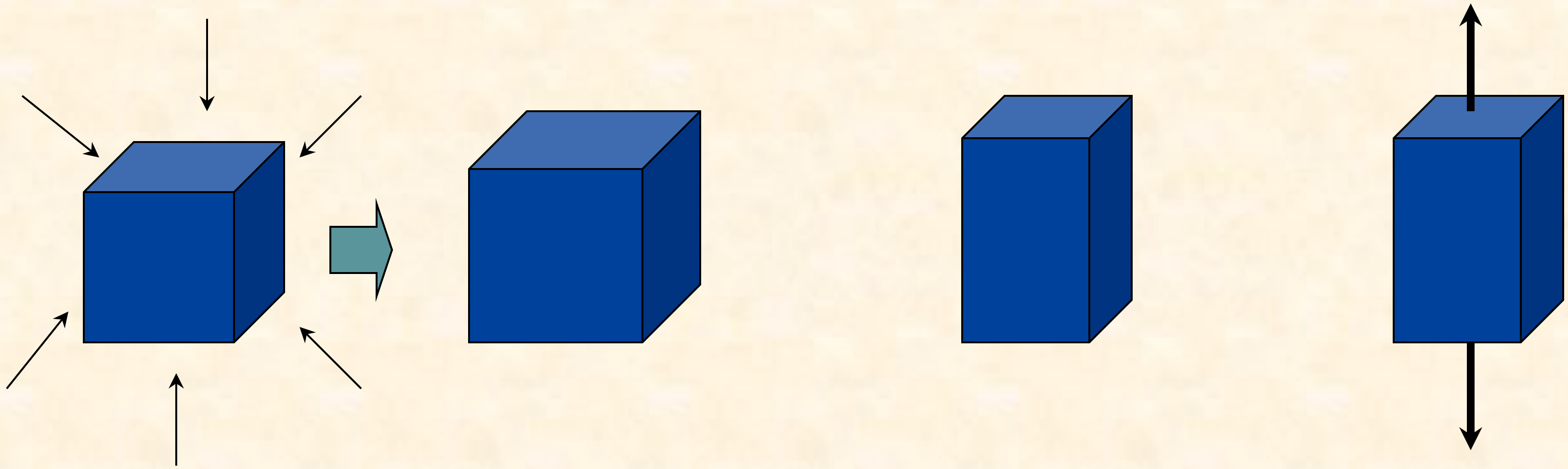
Other forms of degradation that don't fit above, but are still really important!

Localized Deformation Occurs In Many Irradiated Material Systems



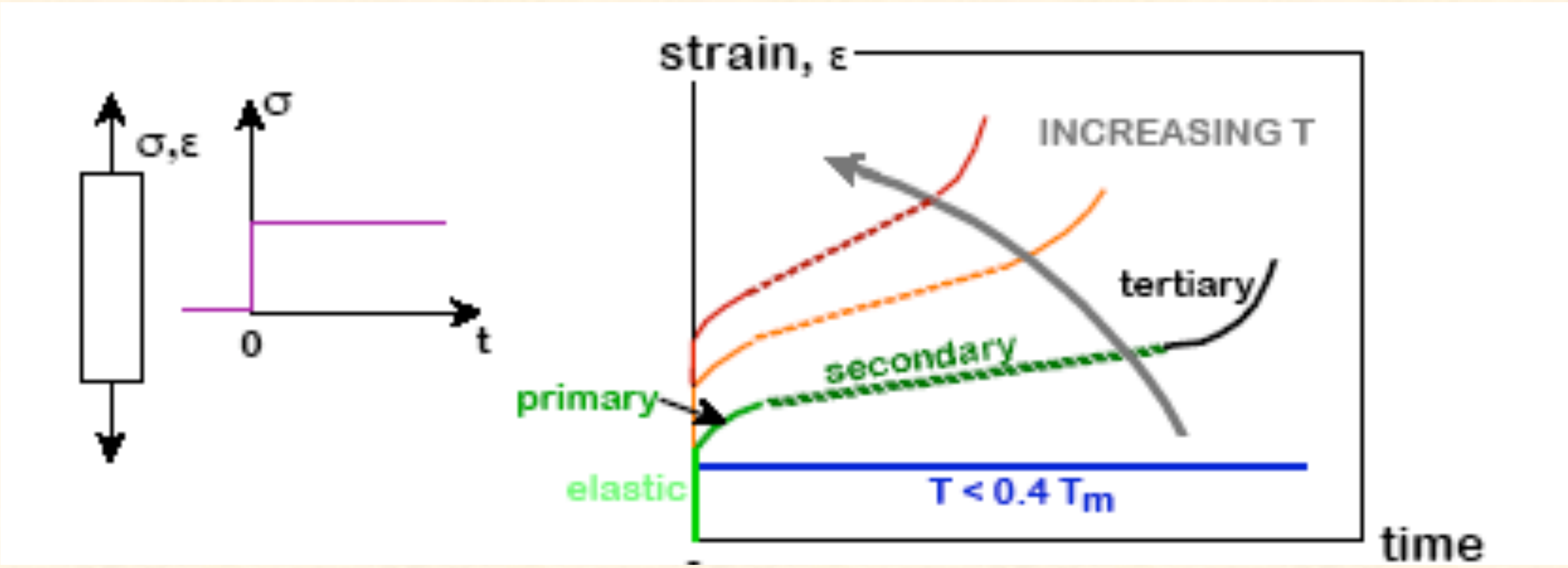
Dimensional stability is heavily influenced by irradiation

	swelling	growth	creep



	unstressed	unstressed	stressed
	$\Delta V/V > 0$	$\Delta V/V = 0$	$\Delta V/V = 0$

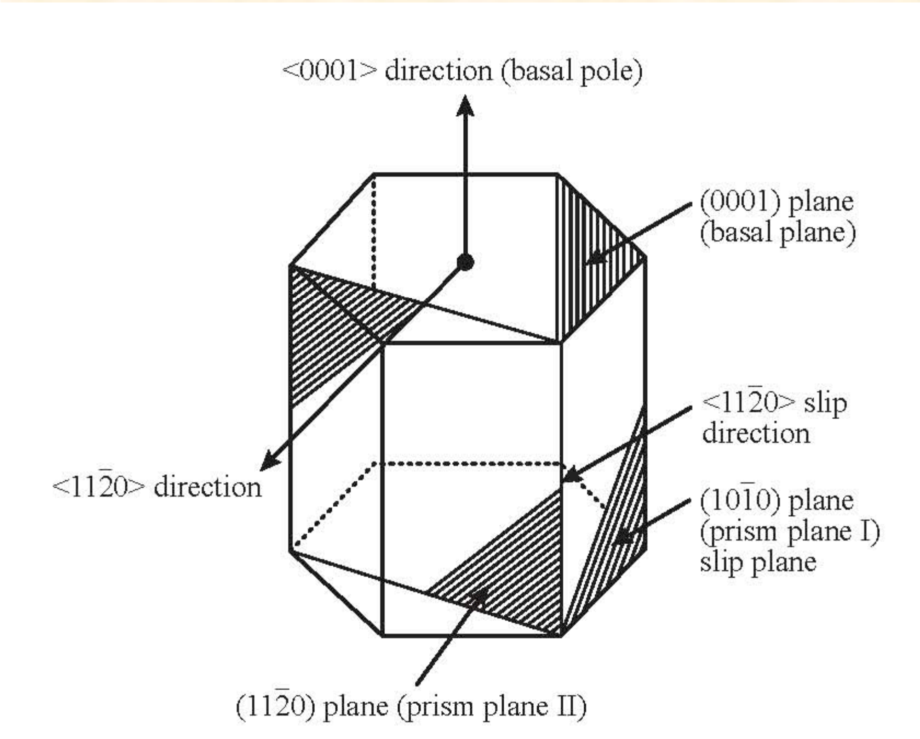
Irradiation also influences creep processes

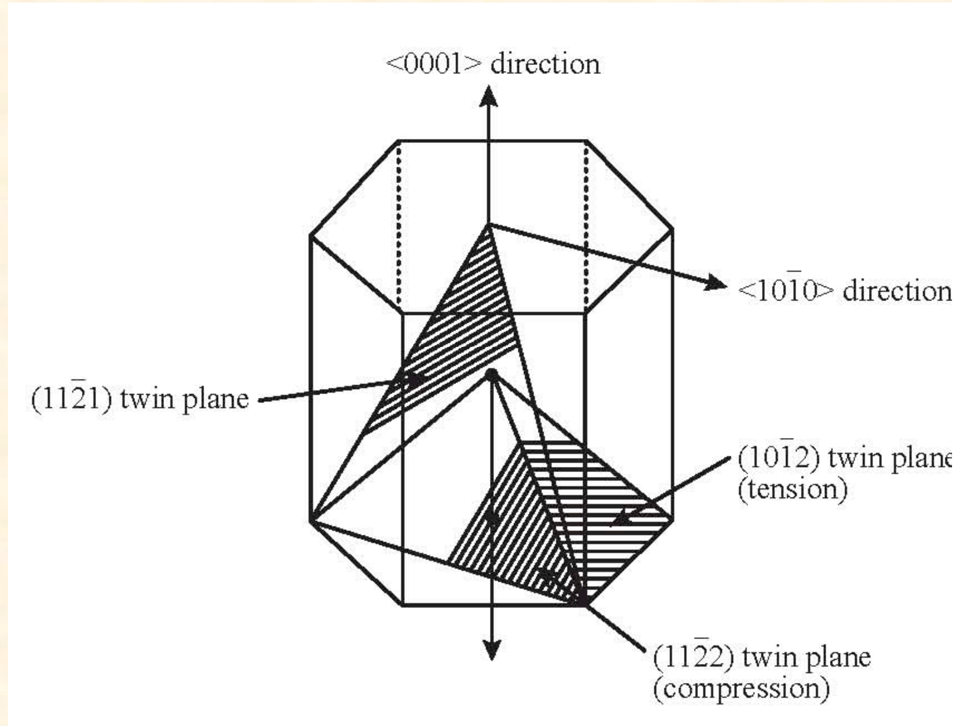


- **Radiation produced point defects increase diffusion and allow creep at lower temperatures**

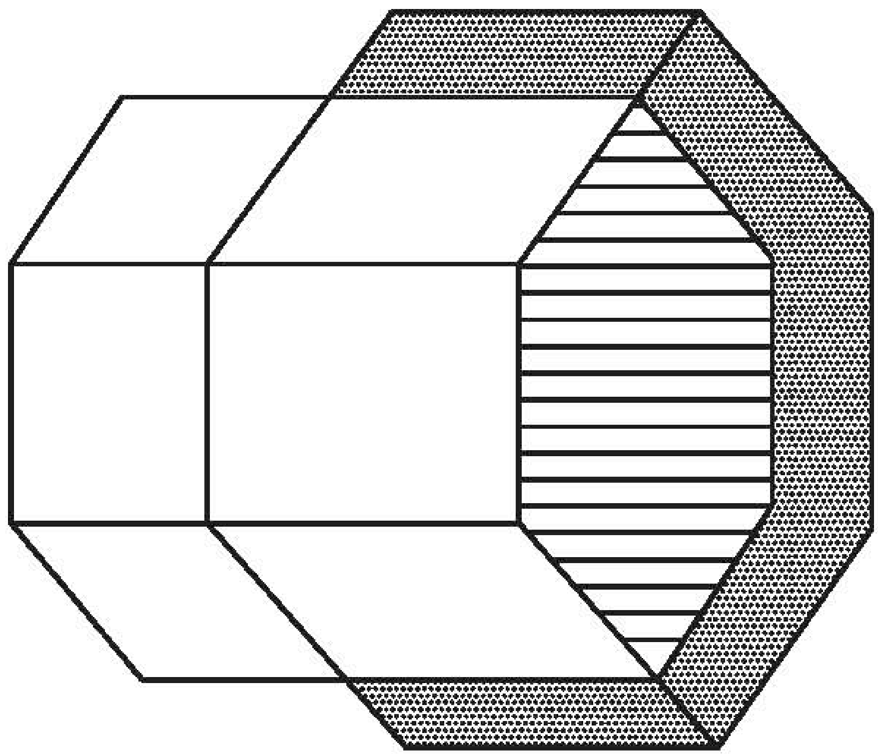
Source: T.R. Allen

Growth is a common issue in Zr-alloys

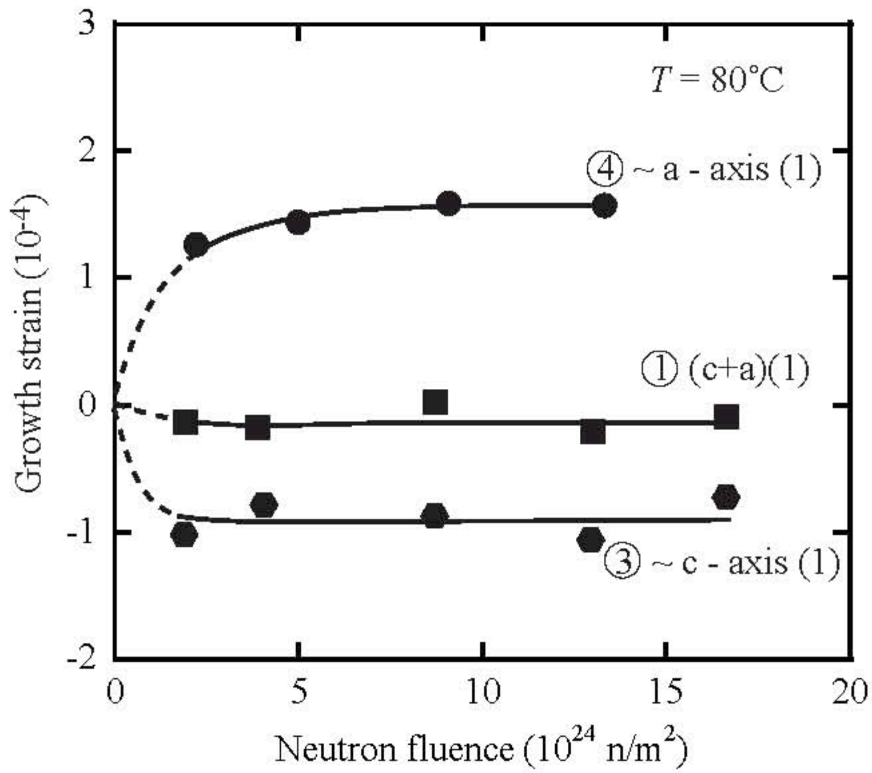




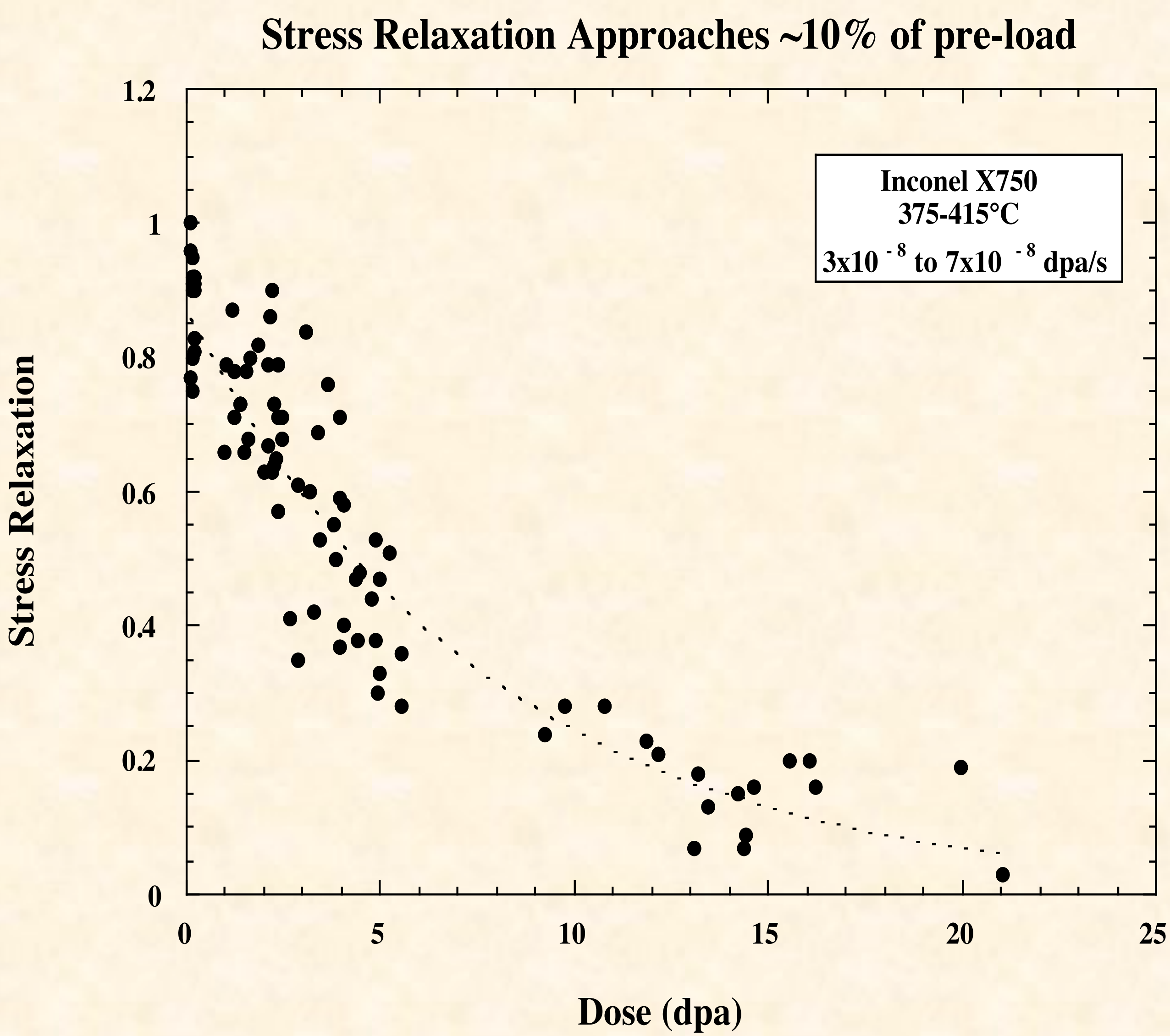
Growth is preferential in Zr-alloys in basal plane directions.



Growth-induced strains vary greatly depending on orientation



Radiation-induced Stress Relaxation can be an issue for some components

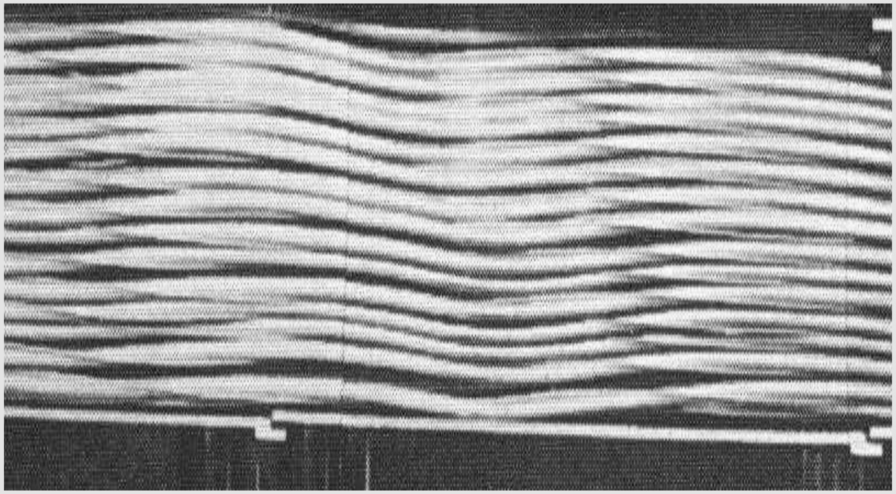


Source: T.R. Allen

Swelling behavior under constrained conditions

- Swelling strains are isotropically distributed in the absence of constraint.
- In presence of constraint the swelling strain is directed toward unconstrained or less-constrained directions.
- Constraints can be externally applied or arise internally from gradients in swelling.
- "Thick vs. Thin" data

"Thin" implies no gradients in temperature, dpa rate or stress.



Stresses generated by swelling or swelling gradients will never exceed the yield stress.

Source: F. Garner

Transmutation

- Irradiation in a neutron environment can also result in transmutation.
- Transmutation products may influence materials properties.
  - Segregation
  - Precipitation
  - Chemical compatibility!

A number of common transmutation reactions in reactors can influence irradiation performance

- A number of important reactions occur in reactor environments, varying with spectrum and materials

- Most create helium
  - $^{58}Ni + n_f \rightarrow {}^{55}Fe + {}^{4}He$
  - $^{60}Ni + n_f \rightarrow {}^{57}Fe + {}^{4}He$
  - $^{58}Ni + n \rightarrow {}^{59}Ni + \gamma \rightarrow {}^{56}Fe + {}^{4}He$
  - $^{10}B + n \rightarrow {}^{7}Li + {}^{4}He$

- He production is of interest due to implications on embrittlement in fast reactors.

Helium Embrittlement for fast reactors

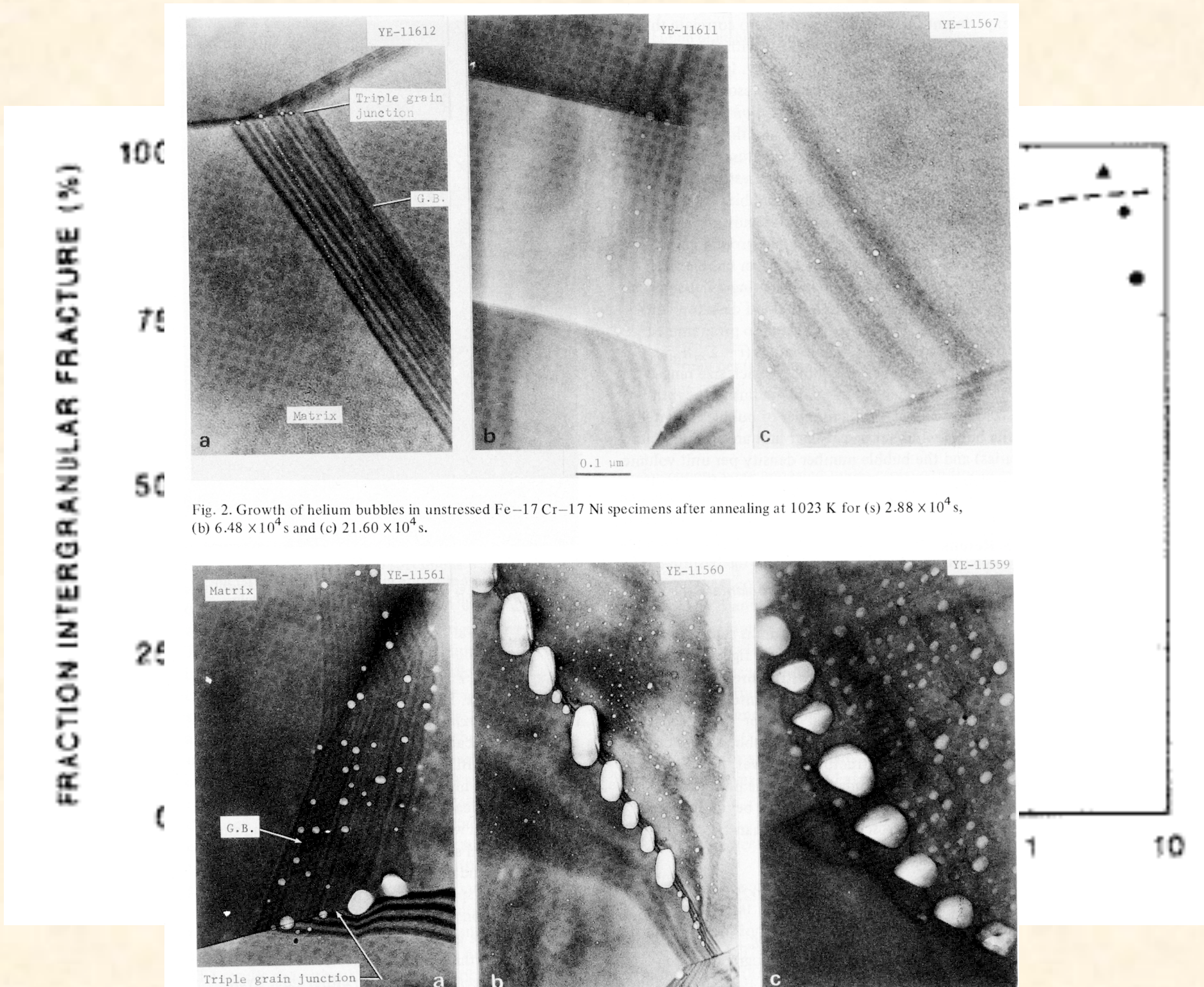


Fig. 2. Growth of helium bubbles in unstressed Fe–17 Cr–17 Ni specimens after annealing at 1023 K for (s) 2.88×10^4 s, (b) 6.48×10^4 s and (c) 21.60×10^4 s.

0 MPa

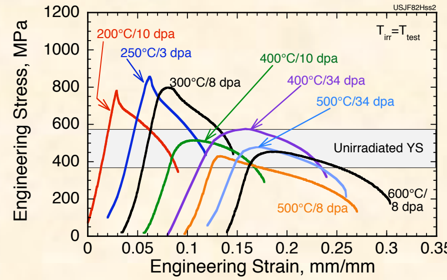
19.6 MPa

Embrittlement via intergranular fracture is dependent on helium content, temperature, and strain rate

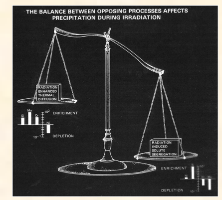
van der Schaaf and Marshall, 1983

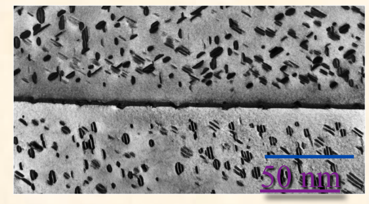
Radiation Damage can Produce Large Changes in Structural Materials

- **Radiation hardening and embrittlement ($<0.4\ T_M$, >0.1 dpa)**



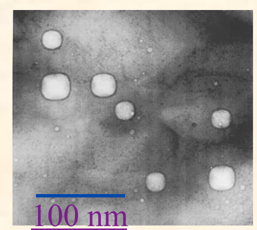
- **Phase instabilities from radiation-induced precipitation ($0.3\text{-}0.6\ T_M$, >10 dpa)**

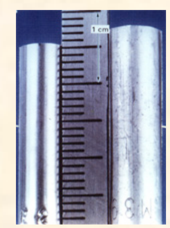




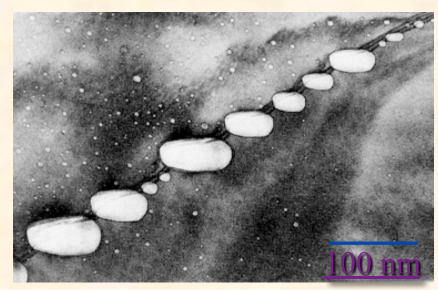
- **Irradiation creep ($<0.45\ T_M$, >10 dpa)**

- **Volumetric swelling from void formation ($0.3\text{-}0.6\ T_M$, >10 dpa)**



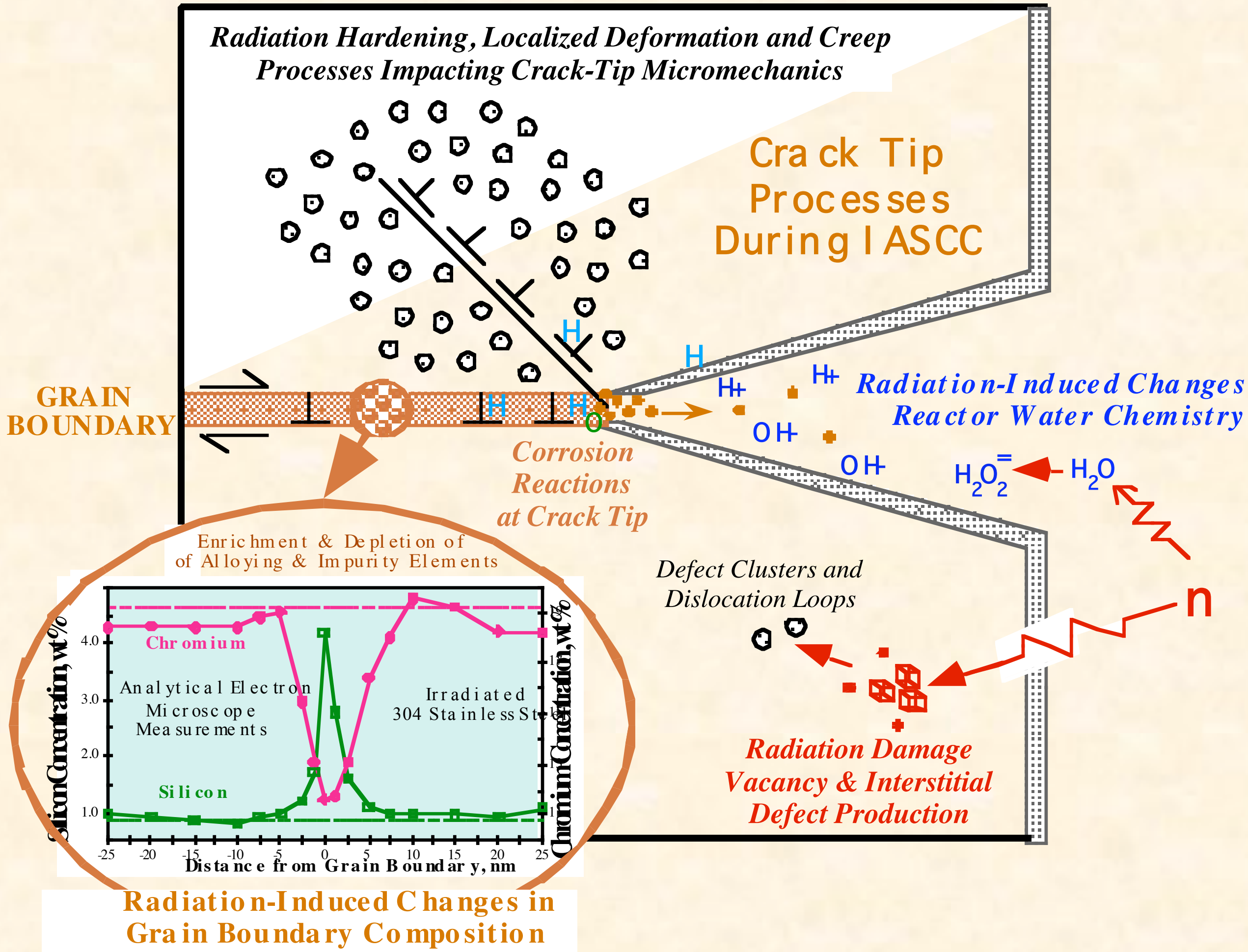


- **High temperature He embrittlement ($>0.5\ T_M$, >10 dpa)**



Source: S. Zinkle

Effects of Irradiation also influence corrosion processes



S. Bruemmer and E. Simonen, Corrosion 50 (1994) 940.

Summary

- All forms of irradiation damage are related to the creation and elimination of excess vacancies and interstitials.
- The interactions between these vacancies and interstitials determine the dominant form of irradiation-induced degradation.
- Many factors are important (temperature, dose, dose rate, alloy condition, alloy, stress and stress state).
- In LWR's, several forms of degradation are observed (RIS, RIP, hardening, embrittlement, swelling and creep)
- Other forms of degradation occur in other reactor applications.

Thanks to the following for allowing me to use their slides!

- Gary Was
- Todd Allen
- Randy Nanstad
- Steve Zinkle
- Everett Bloom
- Steve Bruemmer
- Frank Garner
- Roger Stoller
- Lance Snead